

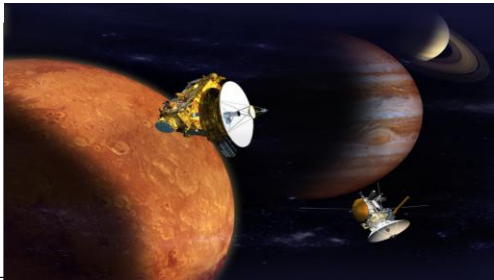
# NIAC Phase I Final Report

## Economical Production of Pu-238

Contact PI: Dr. Steven D. Howe  
Center for Space Nuclear Research (USRA) - non-profit  
phone: (208)526-6103  
email: showe@csnr.usra.edu

Co-Is: Dr. Douglas Crawford, Mr. Jorge Navarro, Dr. Robert C. O'Brien, Jeff Katalenich  
Center for Space Nuclear Research (USRA)

Co-I: Dr. Terry Ring  
University of Utah

<p style="text-align: center;"><b>NASA NIAC Phase II</b> <b>Economical Production of Pu-238</b> PI: Dr. Steven D. Howe Center for Space Nuclear Research showe@csnr.usra.edu</p>	
<p><u>Identification and Significance of Innovation</u></p> <ol style="list-style-type: none"><li>1) All space exploration missions traveling beyond Jupiter must use radioisotopic power sources for electrical power.</li><li>2) The best isotope to power these sources is plutonium-238. The US supply of Pu-238 is almost exhausted and will be gone within the next decade.</li><li>3) The Center for Space Nuclear Research has conceived of a potentially better process to produce Pu-238 than the historic DOE process.</li><li>4) The CSNR method may allow Pu-238 to be available years earlier and 10s of millions of dollars cheaper.</li></ol>	
<p><u>Technical Objectives</u></p> <ol style="list-style-type: none"><li>1) Validate the production rate of Pu-238</li><li>2) Demonstrate the ability to mechanically move sufficient mass through the feed-line</li><li>3) Demonstrate the ability to efficiently extract the produced plutonium</li><li>4) Demonstrate the ability to fabricate spheres of Pu and sinter the spheres into a pellet consistent with current MMRTGs</li><li>5) Demonstrate that the process produces a significantly reduced waste stream .</li><li>6) Determine the cost of a private venture supported production facility</li><li>7) Experimentally answer all major issues to allow engineering design of the facility to proceed via private or government means</li></ol>	<p><u>NASA Applications</u></p> <ol style="list-style-type: none"><li>1) Enable all missions past Jupiter</li><li>2) Enable small satellites, i.e. Cubesats, to be launched to deep space</li><li>3) Enable mobile "hoppers" to be developed for planetary exploration</li></ol> <p><u>Non-NASA Applications</u></p> <ol style="list-style-type: none"><li>1) The method proposed may allow private to build and operate the facility</li><li>2) A low mass, high efficiency power source may enable the NSF Ocean Observation Initiative place sensor arrays and mobile sensors throughout the oceans.</li></ol>

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## **Economical Production of Pu-238**

### **I. Executive Summary**

All space exploration missions traveling beyond Jupiter must use radioisotopic power sources for electrical power. The best isotope to power these sources is plutonium-238 (Pu-238). The US supply of Pu-238 is almost exhausted and will be gone within the next decade. The Department of Energy has initiated a production program with a \$10M allocation from NASA but the cost is estimated at over \$100 M to get to production levels. The Center for Space Nuclear Research has conceived of a potentially better process to produce Pu-238 earlier and for significantly less cost. Potentially, the front end capital costs could be provided by private industry such that the government only had to pay for the product produced.

In the Phase I NIAC grant, the CSNR has evaluated the feasibility of using a low power, commercially available nuclear reactor to produce 1.5 kg of Pu-238 per year. The impact on the neutronics of the reactor have been assessed, the amount of Neptunium target material estimated, and the production rates calculated. In addition, the size of the post-irradiation processing facility has been established. Finally, as the study progressed, a new method for fabricating the Pu-238 product into the form used for power sources has been identified to reduce the cost of the final product. In short, the concept appears to be viable, can produce the amount of Pu-238 needed to support the NASA missions, can be available within a few years, and will cost significantly less than the current DOE program.

The alternative method to produce Pu-238 is to continuously flow an encapsulated aqueous solution containing a high concentration of dissolved Np-237 in a water carrier stream. Once the optimum irradiation period is completed, the encapsulated target slowly moves through the reactor's water tank which allows for time for the decay of Np-238 to Pu-238. This process allows small quantities of Pu-238 to be processed on a weekly basis so that a much smaller, and less costly, facility is needed to accumulate the Pu-238. One other aspect that has come out of the Phase I effort is the recognition that the new process will produce a substantially smaller waste stream of radioactive acidic solution, mixed waste, which has to be stored or processed. Thus, the method will produce substantially reduced costs.

In addition to the technical assessment of the production, the study sought to determine the answers to two major issues: 1) given a sufficient price per kg of Pu-238, could a sufficient return on investment (ROI) be possible so that private venture would pay the up-front capital costs saving the government this requirement in times of diminished budgets, and 2) is it more cost effective to install the new reactor on private land with private operations rather than locating the reactor at a DOE facility? The results of the study indicate that a 20% ROI is possible if the price per kg paid for the material is commensurate with the last known, circa 2007, asking price from Russia. The results also show that, due to lower security and transportation costs, the only responsible option is to locate the reactor at the Idaho National Laboratory site of the DOE.

The Phase I effort utilized experimentally measured neutron spectra from a 1 MW TRIGA reactor at the Kansas State University to estimate the Pu-238 production. Although several

reactor types may be available for Pu-238 production, the TRIGA was used to model the production rates due to the large database regarding neutron flux and costs. By assuming a linear scaling of the neutron flux but keeping the neutron spectra the same, the production rate of the Pu-238, the concentration of Pu-236, and the amount of fission products could be calculated. The calculations show that an irradiation time of between 15 to 18 days with a 12 day decay time is optimum. Pu-236 contamination should be less than 2 ppm. The amount of fission products is estimated to be 150 gms/yr.

Using an 18 day irradiation time, the production rate versus neutron flux, i.e. reactor power, was determined. The trade studies indicate that a reactor between 3.8 to 10 MW can produce 1.5 to 6.25 kgs/yr of Pu-238 respectively. If a 20% return on investment is required, i.e. if the facility is privately funded, the price of the Pu-238 sold to the DOE would have to be 10 \$/kg and 4.3 \$/kg respectively. If no ROI is required, i.e. the US government funds the facility, then the price is 4.9 \$/kg and 1.6 \$/kg for the 1.5 kg and the 6.25 kg respectively. In either case, the results of this study indicate that the 1.5 kgs/yr of Pu-238 can be produced in a new facility within a 3-4 year timeframe for around \$50M and return a 20 % ROI to an investor group.

The primary issue now facing the project is more mechanical than nuclear. We have computationally demonstrated that 1.5 kgs of Pu-238 can be produced annually in a modest sized, commercially available nuclear reactor. The cost of the system will be substantially less than the estimated cost of the DOE program using government owned, high power reactors. The next question to be answered is whether a sufficient amount of Neptunium laden capsules can be pushed through the feedline around the reactor core. A lower power reactor will cost less but needs more Neptunium surrounding it. A higher power reactor needs less Neptunium and produces less waste but will initially cost more. To take the project to the point of gaining an Authority to Proceed decision, the following should be completed: 1) experimentally construct the feedline and determine the maximum mass of Neptunium that can be passed through the system, 2) computationally model an altered design of a commercial reactor to increase Pu-238 production, 3) demonstrate the efficiency of the extraction process using nonradioactive surrogates, and 4) demonstrate the ability of fabricate an RTG pellet with appropriate physical characteristics using spheres as a feedstock. Completion of these tasks will enable the concept to be ready for implementation.

## II. Introduction

Every mission launched by NASA to the outer planets has produced unexpected results. The Voyagers I and II, Galileo, and Cassini missions produced images and collected scientific data that totally revolutionized our understanding of the solar system and the formation of the planetary systems. These missions have been enabled by the use of Plutonium-238 (Pu-238). The conversion of the radioactive decay heat of the Pu-238 to electricity provides a long lived source of power for instruments. Unfortunately, the supply of Pu-238 is about to run out. Developing a reliable supply of Pu-238 is crucial to almost all future space missions.

With current NASA mission plans, all of the Pu-238 remaining on Earth will be consumed within the next decade. After this mission, no spacecraft will travel beyond Jupiter or within the orbit of Mercury. Currently, the US government has initiated a plan to start production of Pu-238 by using reactors at DOE National Laboratories. Realistic estimates place possible production at 1.5 kg/yr. With the increased pressure on the US Congress to cut spending, the Pu-238 production program may slow or be halted entirely.

Radioisotopic Thermoelectric Generators (RTGs) have been used in the past for all missions past Mars to provide electrical power to the platform. The upcoming Mars Science Laboratory, however, will utilize Multi-Mission RTGs (MMRTGs) which can operate in the vacuum of space or in a planetary atmosphere. Because of the desire for no moving parts, reliability, and long life, these systems rely on thermocouples to convert heat to electricity and are inherently inefficient. Only about 6% of the thermal energy is converted into electricity. Consequently, RTGs and MMRTGs require large masses of Pu-238 to provide the heat to make the electricity. In addition, the specific masses of the RTG and MMRTG are 200 kg/kWe and 357 kg/kWe respectively [1]. Thus, the power supplies can be a significant fraction of the platform mass.

Recent advances in Stirling engines at the NASA Glenn Research Center indicate that Advanced Stirling Radioisotope Generators (ASRGs) may provide 25% conversion efficiency [2]. ASRGs will reduce the amount of Plutonium-238 (Pu-238) required for a given power level by a factor of 4. However, ASRGs contain moving parts and may suffer from vibration issues along with shorter life-spans than MMRTGs. In addition, the specific mass of the ASRG is 141 kg/kWe.

Currently, the DOE has proposed to produce Pu-238 using the Advanced Test Reactor (ATR) and the High Flux Isotope Reactor (HFIR) reactors at the Idaho National Laboratory (INL) and Oak Ridge National laboratory (ORNL) respectively. These reactors produce high fluxes of thermal neutrons and are very appropriate for Pu production. Recent estimates of actual production of Pu-238 indicate a rate of 1.5 kg/yr. However, the DOE has estimated that the cost to reach full production will be around \$100M. In 2012, NASA allocated \$10M to DOE to start the production effort.

Current production methods rely on neutron irradiation of large samples of a few kgs of Neptunium-237 (Np-237) for a period of up to one year. The Np-237 will capture a neutron to make Np-238 which decays in 2.117 days to Pu-238. Unfortunately, the Np-238 has a very large fission probability so that around 85% of the Np-238 that is produced is destroyed before it can decay. In addition, after the irradiation, the large sample must be processed for the Pu-238 to be

removed and accumulated. The facility needed to handle large quantities of highly radioactive material is large, complex and costly. In addition, large quantities of radioactive, acidic waste is generated that must be processed or stored for long periods.

An alternative method is to continuously flow an encapsulated aqueous solution containing a high concentration of dissolved Np-237 in a water carrier stream. The use of discrete capsules will make the separation process safer, cleaner and the sampling process more efficient. The capsule (made of one of several viable polymers) also provides another layer of thermal moderation to take advantage of the high thermal absorption cross section of  $^{237}\text{Np}$ . In addition, if there is a pipe break in the water stream carrier, the capsules are easily retrieved and the reactor is not contaminated by the water stream which means there will be no change in reactivity in the nuclear reactor.

Once the irradiation period is completed, the encapsulated target slowly moves through the reactor's water tank and allows time for the decay of Np-238. Because the target is encapsulated, the isotopic concentration can be identified with various radiation spectrometers before the separation column steps. If the product does not contain the desired isotopic concentration, the capsule can be rerun through the nuclear reactor. After irradiation, the capsule contents are individually run through a standard ion exchange column to remove the Pu-238 specifically. This is the same process currently used by the DOE but at a smaller scale allowing for smaller and a less costly separation facility. This process allows small quantities of Pu-238 to be processed on a weekly basis so that a much smaller, and less costly, facility is needed to accumulate the Pu-238.

During the Phase I effort, we recognized that the new process will produce a substantially smaller waste stream of radioactive acidic solution, i.e. mixed waste, which has to be stored or processed. The current method produces 1000s of gallons of liquid waste per year. The new method will produce a few kgs of resin beads covered with grams of fission products. Thus, the method will produce substantially reduced costs.

During the Phase I study, we also recognized another possible cost reduction to the overall process of making sintered pellets for the MMRTGs, i.e. the "back end" of the process. The current process uses solid Pu-238 and ball-mills the material to make a distribution of powder. Some of the particles are sub-micron in size. These small particles migrate through seals in glove boxes and are responsible for all of the worker exposures over the past few decades. The CSNR has identified a potential process that will accept the aqueous solution resulting from the production process and produce large diameter spheres of Pu-238. Compaction of the spheres into a standard "pellet" geometry with the correct physical properties would enable less handling by human workers, a reduced facility footprint, reduced cost, and a smaller waste stream.

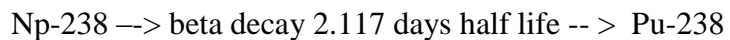
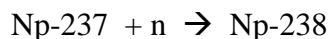
The CSNR has evaluated the potential performance of a continuous flow system by: 1) determining the maximum concentration of Np-237 that can be present in the capsules without suppressing the neutron flux too far, 2) determining the optimum residence time in the reactor to maximize Pu-238 production, 3) predicting the impact of cooling the flow stream to increase the neutron absorption, 4) designing post-irradiation separation chemical process of Pu-238 and 5) evaluating any chemical reactions, erosion, or out-plating of the Np in the system.

In addition, the Phase I project is supporting experimental efforts to 1) demonstrate the fabrication of oxide spheres from aqueous solution using depleted uranium-238 as a surrogate for the plutonium-238, and 2) conduct a proof of concept irradiation experiment to validate the computational models. The experiment will irradiate a sample of neptunium to validate the production rate.

The primary issue now facing the project is more mechanical than nuclear. We have computationally demonstrated that 1.5 kgs of Pu-238 can be produced annually in a modest sized, commercially available nuclear reactor. The cost of the system will be substantially less than the estimated cost of the DOE program using government owned, high power reactors. The next question to be answered is whether a sufficient amount of neptunium laden capsules can be pushed through the feedline around the reactor core. A lower power reactor will cost less but needs more Neptunium surrounding it. A higher power reactor needs less Neptunium and produces less waste but will cost more. To take the project to the point of gaining an Authority to Proceed decision, the following should be completed: 1) experimentally construct the feedline and determine the maximum mass of Neptunium that can be passed through the system, 2) computationally model an altered design of a commercial reactor to increase Pu-238 production, 3) demonstrate the efficiency of the extraction process using nonradioactive surrogates, and 4) demonstrate the ability of fabricate an RTG pellet with appropriate physical characteristics using spheres as a feedstock. Completion of these tasks will enable the concept to be ready for implementation.

## II.a. Concept Description

The process proposed by the DOE for Pu production is a proven method which has been used in the past. In essence, long aluminum tubes are filled with solid neptunium dioxide powder, 20 volume %, and placed into the HFIR or ATR reactors for over 6 months. Pu-238 is produced after neutron capture in the Np-237 via reactions:



The problem with this process is that the Np-238 has a drastically large probability of fissioning before it decays. Thus, for long irradiations, around 80-90% of the Np-238 is destroyed before it can decay. This means that the targets have a large inventory of fission products making them highly radioactive and very hard to handle and process. Consequently, the facility necessary to handle several kgs of highly radioactive Np is large and expensive. In addition, a large waste stream of highly radioactive solution is produced that must be stored or processed. According to R. M Wham of the Oak Ridge National Laboratory in a paper presented at the Lunar and Planetary Science Conference, March 2011, “thousands of gallons of radioactive, acidic solution and several 5 gallon drums of trans-actinide waste are produced and need to be stored.”

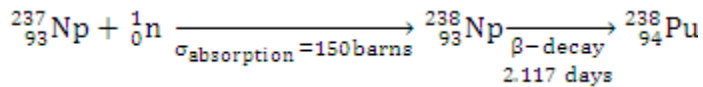
An alternative method that would enable higher production efficiencies of the Pu-238, is to flow the Np in a solution through the core of the reactor and extract the Pu-238 continuously at lower mass rates. This process produces Np-238 and then removes it from the reactor before substantial fraction can burn up and be lost. It also allows for much smaller processing facility because smaller amounts are processed continuously and the material is not full of fission products. The waste stream is, thus, significantly reduced.

The flowing target scenario is not possible to implement in the ATR or HFIR without major interruption of service and extensive cost. However, the flowing target can be implemented in a new, commercially available reactor. While several vendors can supply such a reactor, this study has used the TRIGA reactor available for General Atomics as a test case.

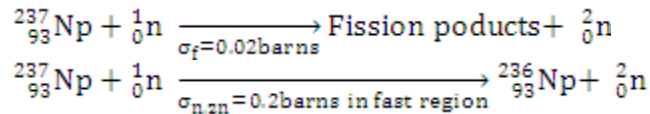
The primary innovation of this proposal is the continuous flow feedstream and the use of capsules to “quantize” the feedstream into small units to be treated individually. The capsules allow a solution with a high concentration of Np-237 to be irradiated without the risk of the Np plating out along the flow channel. Use of the capsules also allows small quantities of irradiated material to be processed at a time which reduces the processing facility’s size even more. Finally, the use of capsules mitigates the risk of having the pipe break and spill the target solution into the reactor or facility. What is required, then, is a reactor with a high thermal flux of neutrons and the ability to alter its design to accommodate the flow channel. The residence time in the core, the decay time out of the core, and the processing of the irradiated solution must then be determined.

## II.b. Production basics

A continuous flow process to produce Pu-238 will requires a thermal neutron flux to take advantage of the very high thermal absorption cross section ( $\sigma_{\text{absorption}}$ ) in the nuclear reaction



compared to a competing nuclear reactions



The third reaction leads to the production of Pu-236. Pu-236 is a contaminant that must be kept to below a 3 ppm level.

Figure 1 shows the difference in the three cross sections [3] with the plot in red being the absorption cross section for  ${}^{237}\text{Np}$ , green plot is the fission cross section and the blue plot is the n,2n nuclear reaction producing Pu-236. Figure 1 clearly shows the need to have a very low energy, i.e. thermal, flux expose the target material. A heavily thermal flux of neutrons will produce Np-238 but not cause fissions in the Np-237 nor produce Np-236 leading to Pu-236.



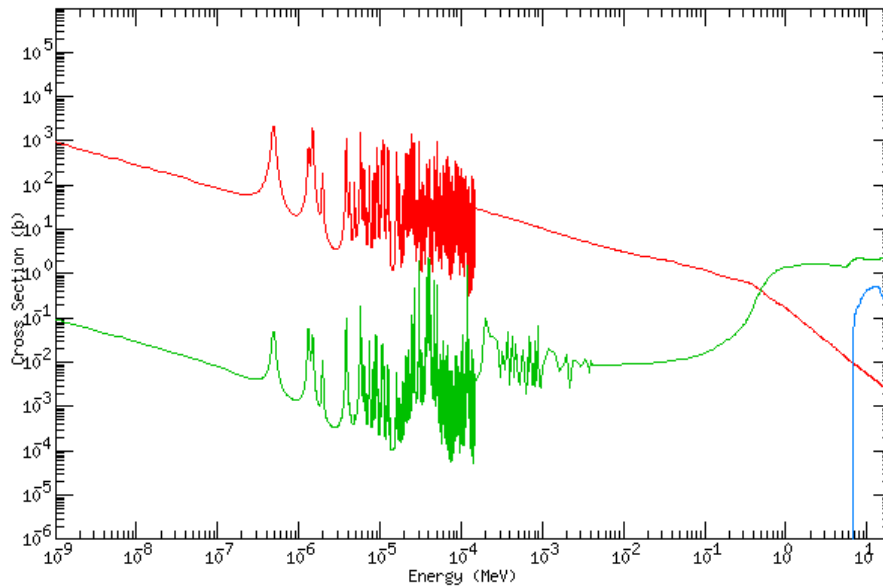


Figure 1. Plot of the energy dependent microscopic cross section, i.e. reaction probability, for Np-237 absorption in red, Np-237 fission in green and Np-237 to Np-236 in blue [3].

Typically, neutrons are born in the fission process at near 1 MeV, clear to the right hand side in Figure 1. Letting the neutrons pass through water allows them to thermalize to energies at the left of Figure 1. However, as the neutrons pass through the water they are absorbed so that the flux level is reduced. Thus, optimization of the Pu-238 production entailed an evaluation of where in the reactor core the target material is placed.

The neutron flux spectra for three different locations in a TRIGA core are shown in Figure 2.

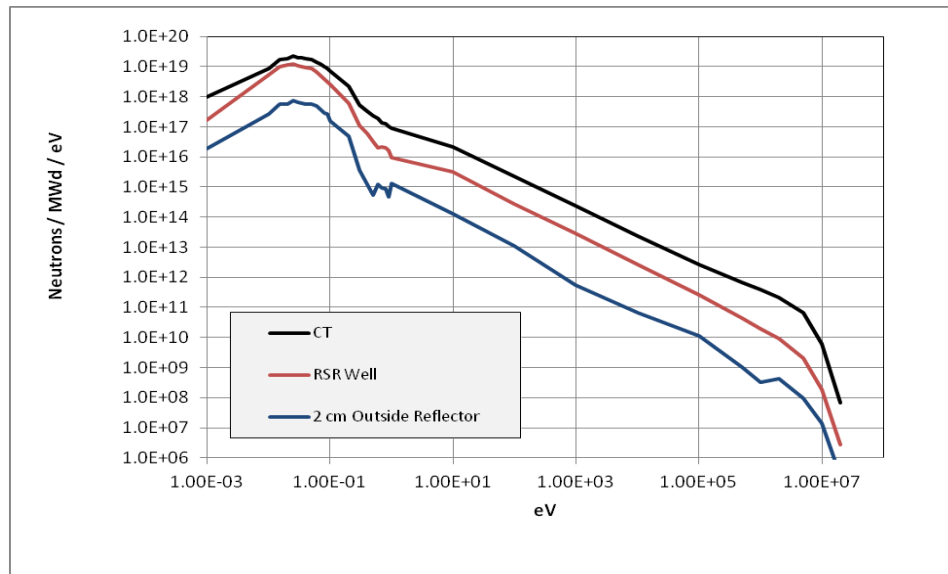


Figure 2. Spectra of neutrons at three locations in the 1 MW TRIGA reactor at Kansas State University.

The spectra show that the neutron fluence outside the core is more thermal, i.e. the ratio of low energy to high energy neutrons is bigger than at the other two locations. However, the magnitude of the flux is lower by two orders of magnitude. Placing the target material in the CT location will breed more Pu-238 by also more Pu-236. Based on these data, the optimum position in the TRIGA core appears to be at the RSR location. Creating such a location in a 5 MW-scale core may require some slight alteration in the commercially available design.

The results showing the time dependence of the production of various isotopes are shown in Figure 3. A key observation from the figure is that the Np-238 reaches an equilibrium level very quickly. In other words, by irradiating the samples longer than a few hundred hours just burns up the Np-238 and increases the fission product concentration. This indicates that the irradiation times should be much shorter than the six months currently employed by the DOE program.

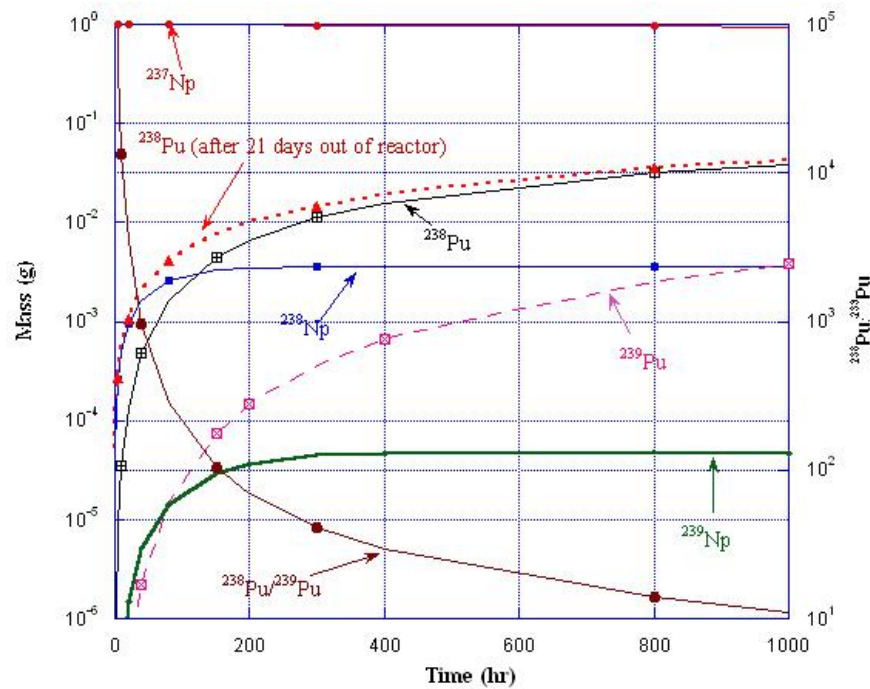


Figure 3. Calculated concentrations of various isotopes as a function of irradiation time in a flux of  $1 \times 10^{14}$  n/cm<sup>2</sup>-s. The masses plotted are per gram of Np-237.

The Phase I study is divided into major sections: 1) production - modeling the neutronics reactions to estimate the amounts of various isotopes made in the reactor, 2) reactor design – modeling the impact on the reactor of having neptunium near the core and impact on the neutron flux of various reactor geometries, 3) system studies – modeling the production line in the reactor and estimating masses and capsule cycling rates, 4) separation chemistry – design of the line to separate the Pu-238, Np-237, and fission products from the nitric acid, and 5) costs – costs of the reactor, the separation line, security and operations.

### III. Results of the Phase I project - modeling

#### III.a. Production modeling

A major focus of Phase I was on assessing the neutronics issues inherent in placing a large quantity of material in a commercial nuclear reactor core. The Np-237 used to breed the Pu-238 absorbs neutrons readily but also produces fission which makes neutrons. The overall impact on the core had to be assessed.

Previous calculations showed that placing several kilograms of Np-237 within a TRIGA core actually shut the reactor down, i.e. made the core sub-critical. One of the first tasks was to reassess this conclusion using the geometry of the continuously flowing feedline around the outside of the core. Calculations using MCNP and Scale validated the idea that several kilograms of Np in solution can be placed around the core with negligible impact on the reactivity.

The next issue was whether a low power reactor had sufficient neutron flux to produce the amount of Pu-238 required without producing too much Pu-236. To produce Pu-238 in a flow loop around a nuclear reactor requires the coordination of several process components for the flowing target stream within a nuclear reactor. This report focuses on a few of these key process components: what neutron spectrum in a nuclear reactor will provide the best product stream (produce Pu-238 with the least amount of other isotopes), what is the optimum irradiation time the target stream should be in the neutron flux to make the 1.5kg/yr of Pu-238 and size of the flow tubes/capsules for the Np-237 dissolved in HNO<sub>3</sub>.

The first key is to determine a location within a nuclear reactor that provides a neutron flux spectrum with a high thermal component. A high thermal flux takes advantage of the very high microscopic thermal absorption cross section ( $\sigma_{\text{absorption}}$ ) of Np-237 which produces Np-238, and then beta decay of Np-238 to produce Pu-238. The second key is to remove the Np-238 from the thermal neutron flux at an optimum time before Np-238 fissions or is activated to Np-239. The last item is how large of a flow tube (diameter of the tube) can be used to maintain the order of magnitude the flux needs to be to activate Np-237 atoms into Np-238 atoms. All three of these components are considered and are presented in this analysis to maximize the production of Pu-238 while minimizing the production of other isotopes via competing nuclear reactions. To determine these process components: a 15-group neutron activation analysis (NAA) was performed with neutron spectra from three different locations in the Kansas State University (KSU) nuclear reactor.

The three locations in the KSU reactor are the central thimble, RSR Well position and 2cm outside of the reactor's reflector. The central thimble is at the center of the reactor, the RSR Well position is on the outside of the reactor core and the last position is 2cm outside of the graphite reflector. More information about the KSU nuclear reactor facility can be found at this link: <http://www.mne.ksu.edu/research/centers/reactor>.

## 15-group Neutron Activation Analysis within the KSU nuclear reactor

All three locations investigated provide a reasonable thermal flux component which is needed to take advantage of the high microscopic thermal absorption cross section ( $\sigma_{\text{absorption}}$ ) of Np-237. Each location has differences through the fast and resonance regions of the neutron spectrum that separate the three choices significantly. The major differences between the three locations are seen by comparison of the following criteria: irradiation time required to make 1.5kg/yr Pu-238, amount of Pu-236 produced via a competing nuclear reaction and number of fissions which is directly proportional to the number of fission products produced instead of Pu-238. The flux spectra in Figure 4 are: normalized to one as the peak value, divided by change in lethargy ( $\Delta u$ ) and plotted versus energy (eV) to show the differences in the three locations spectrums. The three spectra are typical flux spectra in a TRIGA style nuclear reactor.

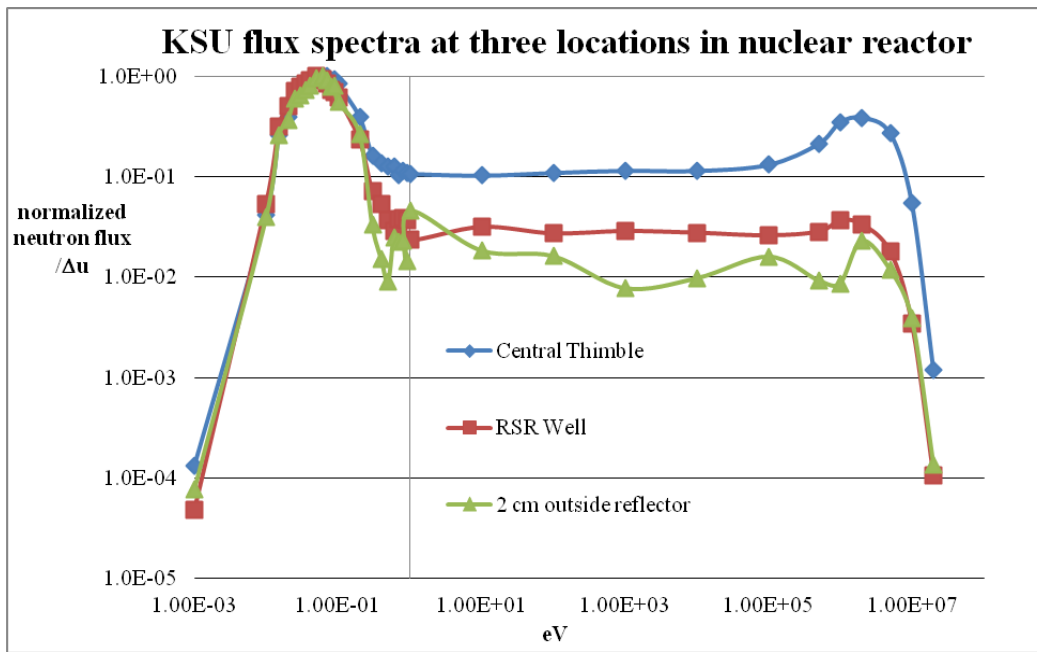
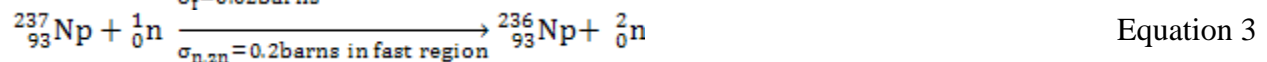
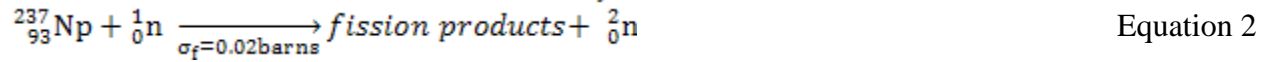
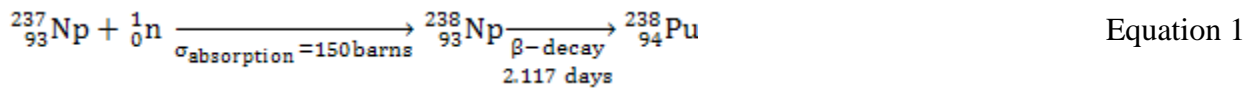
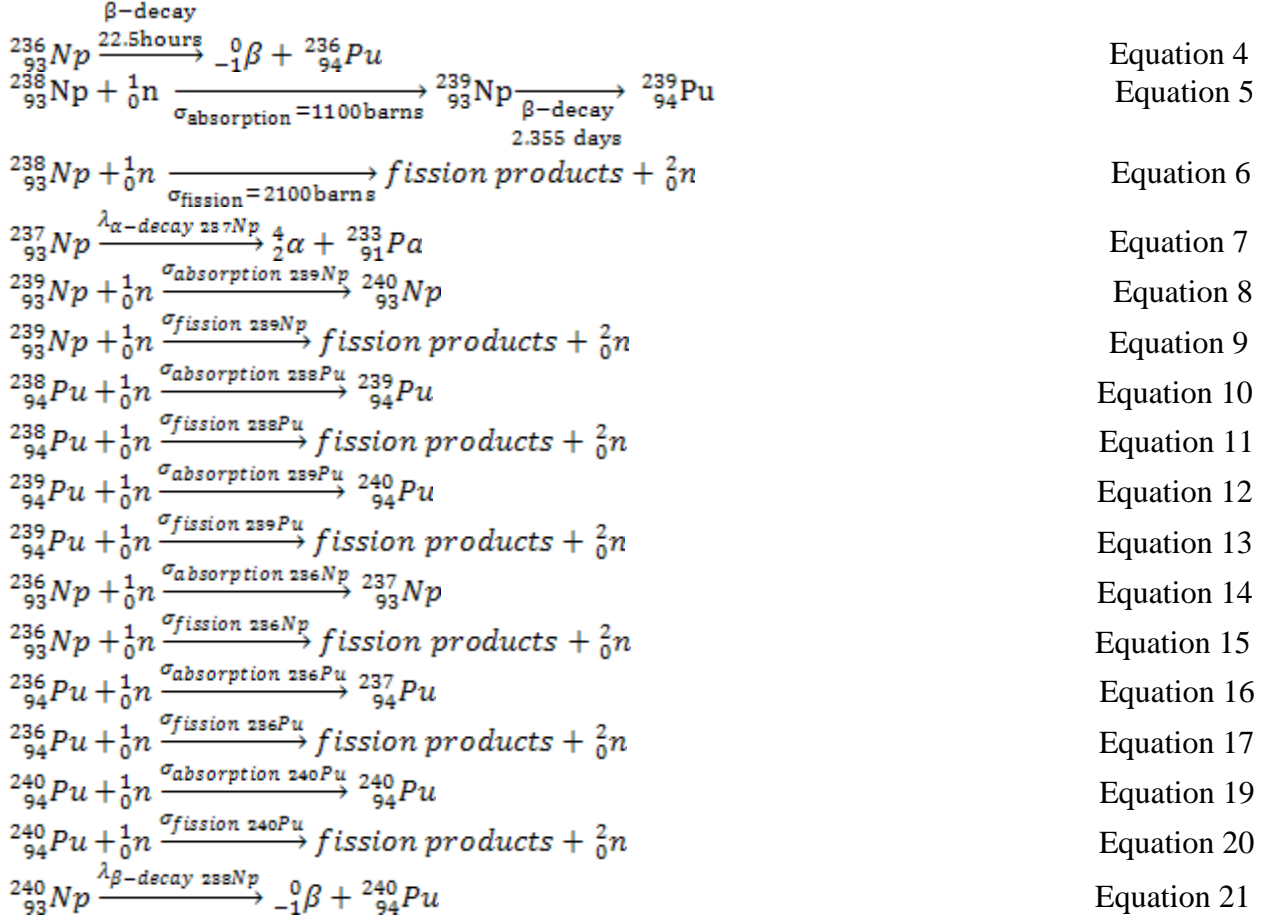


Figure 4 flux spectra for the three locations in the KSU nuclear reactor

To analyze which spectrum produced the best results a set of ordinary differential equations (ODE) based on the nuclear reactions in equations 1-5 were solved with a Runge-Kutta 5,6 numerical approach. Equation 1 is the nuclear pathway that must be optimized to produce the most Pu-238. Equations 2-6 are competing nuclear reactions that need to be minimized. Other nuclear reactions were also modeled to account for their effects in the product stream. Equations 8-21 show these nuclear reactions. Also the presence of Pa-233 from  $\alpha$ -decay of Np-237 is analyzed in the feed stream, see equation 7.





The high absorption cross section for Np-237 is advantageous if there is a thermal flux which all three locations have but all three locations have a fast flux component as well which leads to more fission products and more Pu-236. The cross sections for these nuclear reactions were taken from the following websites: <http://t2.lanl.gov/data/endl/endlvii.1-n.html> and <http://atom.kaeri.re.kr/>.

Figure 5 shows the difference in the three cross sections for Np-237. The threshold for the n, 2n reaction (Equation 3) that produces Np-236 is about 6.0 MeV. The cross over point where the fission cross section for Np-237 is greater than the absorption cross section is about 0.5 MeV. Np-238 also has a high fission cross section which is why the removal of Np-238 before it fissions is a critical component to optimize. Figure 5 shows the microscopic cross sections for Np-238.

The 15 group neutron activation analysis provided insight for the desired production of Pu-238, low Pu-236/Pu-238 parts per million (ppm) levels and a low number of fissions. The groups for the analysis are displayed in Table 1. Figure 6 shows the comparison of ppm levels for the three positions. The fission rate comparison is in Figure 7. The fission rate is calculated by summing up the product of the flux, microscopic cross section and the atom densities for each fissile isotope, i.e.  $\sum_i \phi (\sigma_f N)_i$ .

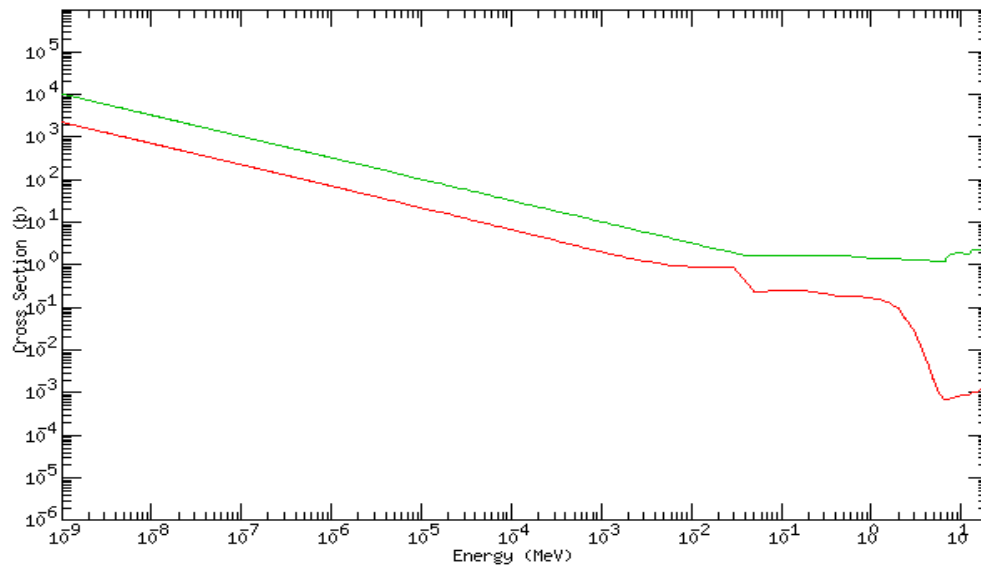


Figure 5.  $^{238}\text{Np}$  fission and absorption energy dependent microscopic cross sections, fission in green and absorption in red

**Table 1** list of energy groups for neutron activation analysis

Group #	MeV
1	0 to 1E-9
2	1E-9 to 1E-8
3	1E-8 to 1E-7
4	1E-7 to 1E-6
5	1E-6 to 1E-5
6	1E-5 to 1E-4
7	1E-4 to 1E-3
8	1E-3 to 1E-2
9	1E-2 to 1E-1
10	1E-1 to 5E-1
11	5E-1 to 1
12	1 to 2
13	2 to 5
14	5 to 10
15	10 to 20

The Neutron Activation Analysis (NAA) was performed assuming a spatially uniform flux with an order of magnitude of  $4 \times 10^{13}$  nts/cm<sup>2</sup>-s which is assumed to relate to roughly a 2.8 MW TRIGA style reactor. This is based on information from the McClellan Nuclear Research Center at the University of California Davis (UC Davis), for more information about the McClellan facility follow this link [http://mnrc.ucdavis.edu/MNRC\\_flux\\_info.pdf](http://mnrc.ucdavis.edu/MNRC_flux_info.pdf). The NAA also assumed 10 kg of Np-237 dissolved into HNO<sub>3</sub> flowing around the reactor for the irradiation time frames. The 10 kg limit is based on the amount of Np-237 that will dissolve into solution which is 432

mg/cm<sup>3</sup> under standard conditions [4], and the surface area of the reactor face for irradiation. From the 432 mg/cm<sup>3</sup> limit a target tube diameter size can be estimated, which is 3 cm.

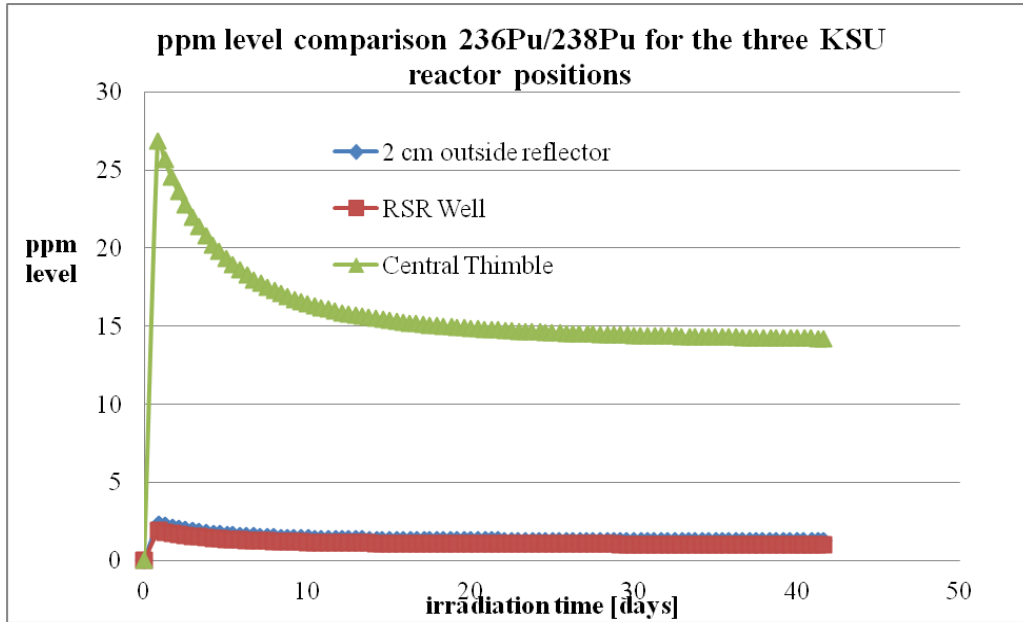


Figure 6. Comparison of the Pu-236/Pu-238 ppm levels of the three spectra in the KSU reactor

The three spectrum locations for these conditions produced different Pu-238 production curves (Figure 8) for the conditions stated. Figure 8 shows the kilograms of Pu-238 that can be produced given the irradiation time on the horizontal axis, production year of 350 days and a 12 day decay time for Np-238 to decay to Pu-238. As an example if the target is irradiated for 20 days in the RSR

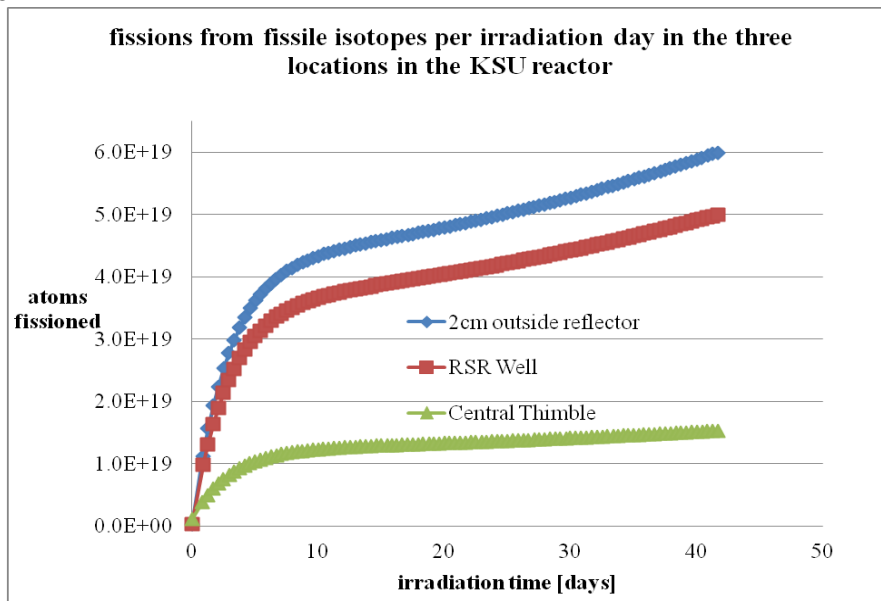


Figure 7. Comparison of the number of fissions in each location in the KSU reactor

Well position then based on a 350 day a year operating cycle and a 12 day decay time frame 2 kg/yr of Pu-238 could be produced in the flowing target process. The production curves in Figure 5 also show that the flowing target process could make more Pu-238 if necessary, a simple shift of irradiation time can be made by slowing down the flow rate and more plutonium could be made.

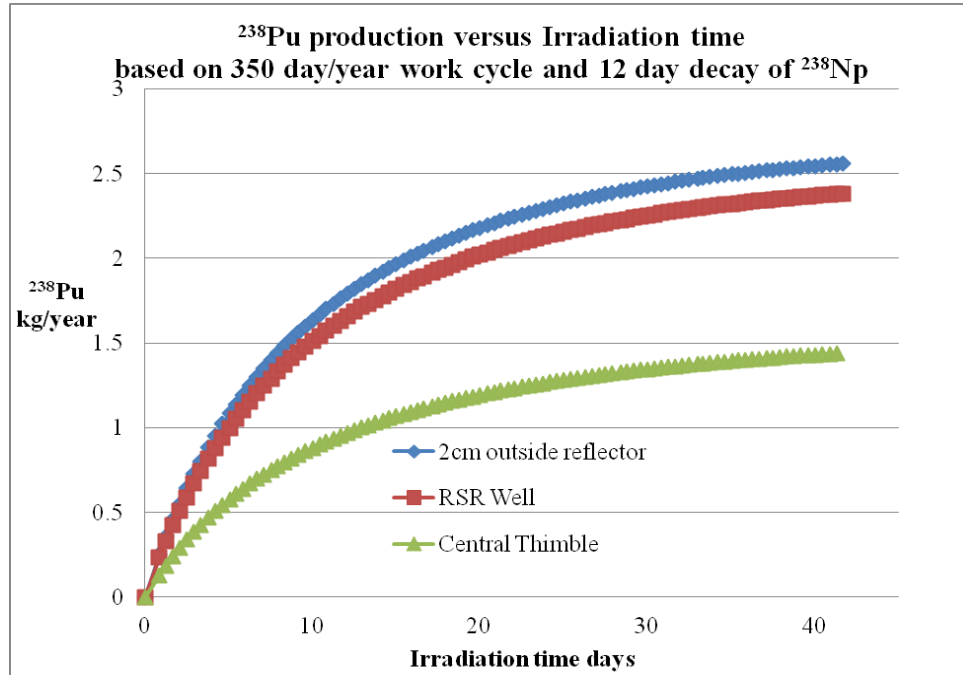


Figure 8. Pu-238 production plot for the three locations in the KSU reactor

The central thimble position spectrum did not produce the 1.5 kg/yr of Pu-238 within the 40 day irradiation time frame of the NAA. This is because at this location the number of fast neutrons was too large and so there was not enough activation of Np-237 to Np-238. At the central thimble location there are less total fissions compared to the other two locations but this is because there is less Np-238 which has a very high fission cross section, see Figure 5. The ppm levels at the central thimble location are excessively high for the ppm specifications (3ppm) of Pu-236 to Pu-238. The excessive ppm levels at the central thimble location are because of the fast neutron flux at this location as well; see nuclear equations 3 and 4. The RSR Well location and 2cm outside of the reflector are similar in terms of the NAA performed, except for the number of fissions produced. The RSR Well position produced less fissions than 2 cm outside of the reflector. The best location for the flowing target is the RSR Well because of issues stated.

The 2<sup>nd</sup> key is the balance of the residence time Np-237 stays exposed to thermal neutrons to become Np-238. The removal of the Np-238/ Np-237 from the neutron flux at an optimum time maximizes the amount of Np-238 that can be extracted and minimizes the amount lost through the competing nuclear reactions shown with nuclear equations 2-7. Np-237 is recycled with this process so a shorter irradiation time can be implemented since the unreacted Np-237 can be sent



back through the reactor loop to produce more Pu-238. The optimum is based on production of Np-238 and this can be seen in Figure 9.

Figure 9 shows the number of atoms produced versus irradiation time in the reactor at the three different locations. The maximum amount of Np-238 made within the constraints of the NAA work is 17 days to 19 days, depending on which location the target is placed. The 1.5kg/yr production rate can be achieved with a shorter irradiation time frame, see Figure 8 but the optimum time for producing the most Pu-238 is a 17 day irradiation for the RSR Well position or 2 cm outside the reflector.

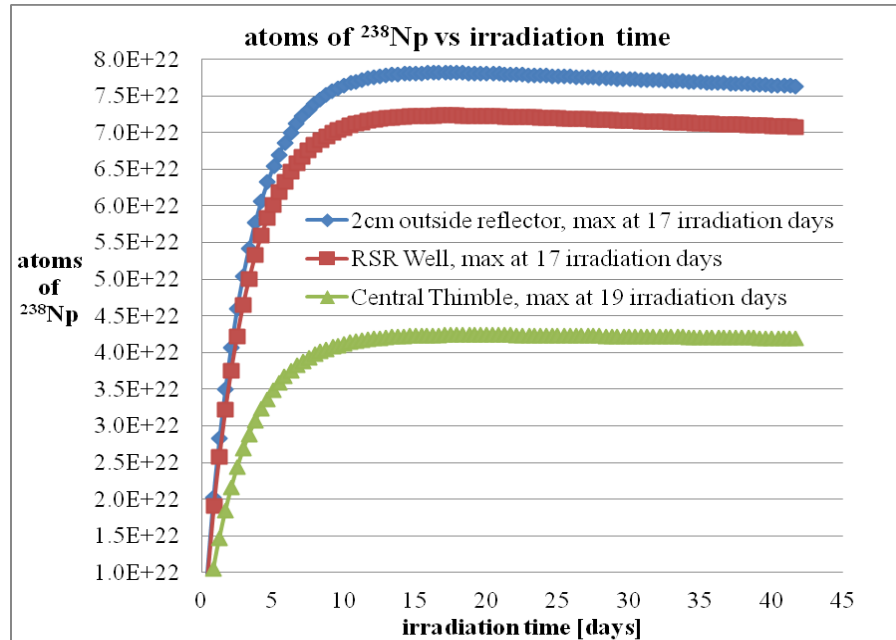


Figure 9. Optimum time for irradiation of flowing Np-237 target

The irradiation time to make 1.5kg/yr of Pu-238 is dependent on the spectrum of the flux as well as the magnitude of the flux. The magnitude of the neutron flux in any reactor is highest at the center which is the central thimble location for the KSU reactor. The magnitude of the KSU reactor was not used for the analysis just the flux spectra at the three locations. The reasoning for using the spectra and not the magnitude is to determine from the NAA the order of magnitude for the flux and the amount of Np-237 dissolved in solution needed to produce the 1.5kg/yr Pu-238 based on the spectra. This method of calculating production with a neutron activation analysis provided insight into where the best location is for placing the flowing reactor target as well as a rough size/cost for the nuclear reactor. From this method, the design of a nuclear reactor can be accomplished in future work that provides the correct order of magnitude, flux spectrum and at the correct location within the nuclear reactor for Pu-238 production.

### III.b. Reactor design

The purpose of this section of the report is to present the results of the methods along with the different simulation models approaches that were implemented to determine the neutronic requirements needed to achieve the desire production of Pu-238. In order to determine those requirements a feasibility study in which different nuclear reactor options were simulated was performed. The main focus of the study was to model different nuclear reactor configurations in order to determine the design parameters needed to make a final recommendation on a reactor design for Pu-238 production using the Np-237 encapsulated target process. Different nuclear reactor simulation approaches were needed to determine the feasibility and requirements of the continuous encapsulated target process due to of the novelty of the method. The approaches used in the past normally placed the Np-237 target inside the reactor core boundaries. This study differs from previous because Np-237 target will be placed in a coil pipe outside boundary of the fuel reactor core. The target position is a significant variation from a material activation stand point. Also having the target outside of the reactor fuel core boundary affects the performance of the reactor and changes the neutronic requirements needed to obtain the desire Pu-238 product. This new Np-237 target configuration has never been model before and even though it seemed like a small change at the beginning of the study, some unforeseen challenges arise and had to be resolved in order to achieve the desire production of Pu-238.

The simulations were performed using the comprehensive modeling and simulation suite for nuclear safety analysis and design SCALE6.1 package. SCALE6.1 contain different sequences for reactor analysis. The reactor analysis and activation Np-237 calculations were performed using the TRITON sequence. The simulations were done using a 238 energy ENDFVII multigroup cross section library that was processed with BONAMI for the unresolved resonance self-shielding calculations, CENTRM for a 1-D point-energy flux calculation, and PMC to use the flux and point cross section data to generate a problem specific multigroup cross sections library.

The initial approach used in this study was to model a generic TRIGA configuration. TRIGA reactors were selected because their core configuration flexibility. In TRIGA reactors, fuel can be shuffled and placed in different positions around the core in order to achieve the desired flux profile at irradiation experiment locations. It was important to start with a generic known reactor in order to learn how the activation of the Np-237 target behaves and to make an initial estimation of flux requirements. Three different reactor configurations each with different modifications were modeled.

#### Model Description and Results

Traditional solid target processes and non-traditional liquid targets processes usually placed the Np-237 target inside experimental traps within nuclear reactor core. The first step of our study was to model a known hexagonal TRIGA reactor. The model is a 1 MW TRIGA core based on the University of Utah reactor [5]. The hexagonal core diameter is 53 cm with a 58.4 cm height. There are 77 twenty percent U-235 enriched zirconium hydride fuel pins with a 3.3 cm diameter and a 38.2 length (Figure 10). There are also some D<sub>2</sub>O water and H<sub>2</sub>O rods along with three

control boron rods (Figure 12). Two different Np-237 target configurations were simulated using the 1 MW TRIGA model.

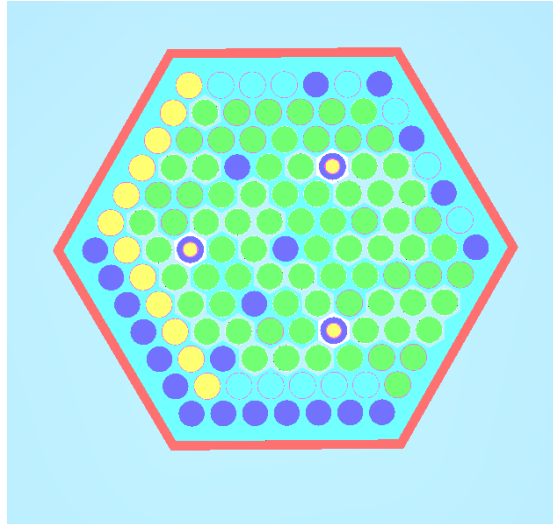


Figure 10.- Cross Section of the Hexagonal Nuclear Reactor Core

The first configuration was model as a tool to learn the limitations and parameters of a traditional process. The second model is the first attempt to place the target outside the fuel core boundaries. For the first 1MW TRIGA configuration the Np-237 targets were placed in different experimental locations inside the core fuel boundary. This configuration simulation of the 1 MW TRIGA reactor determined that the Np-237 encapsulated target when placed inside the reactor acts like a neutron absorber making it difficult to maintain criticality and limiting the amount of Np-237 that can be activated, therefore limiting Pu-238 production.

The second configuration for the 1 MW TRIGA reactor model have the Np-237 target outside the reactor fuel core. This second target configuration of the 1 MW TRIGA was examined because it more closely simulates the desire target placement of having a coil pipe target around the reactor. This second configuration established that placing the Np-237 target outside the reactor fuel boundaries will not decrease criticality; in fact it will act as a reflector. Having the target acting as a reflector signifies that the amount of Np-237 will not negatively affect the performance of the reactor. This discovery was unexpected because the initial thought was that no matter where the Np-237 target was placed it was going to negatively affect criticality, this meant that the amount of Np-237 mass had to be limit in order to avoid shutting down the reactor or having difficulties controlling it. However by placing the target outside the reactor fuel boundaries the critical assembly is not disturb by the introduction of negative reactivity. Having the target outside the reactor actually introduce some positive reactivity into the core however it can easily be accounted for in the final reactor design by changing the worth of the control rods. Figure 11 shows the Np-237 scattering cross section plot against graphite, a well know reflector. The plot demonstrates that the Np-237 is a better reflector than the graphite confirming the findings describe above. The 1 MW TRIGA simulations also conclude that not enough flux was produced to activate the necessary Np-237 to generate the desired Pu-238.

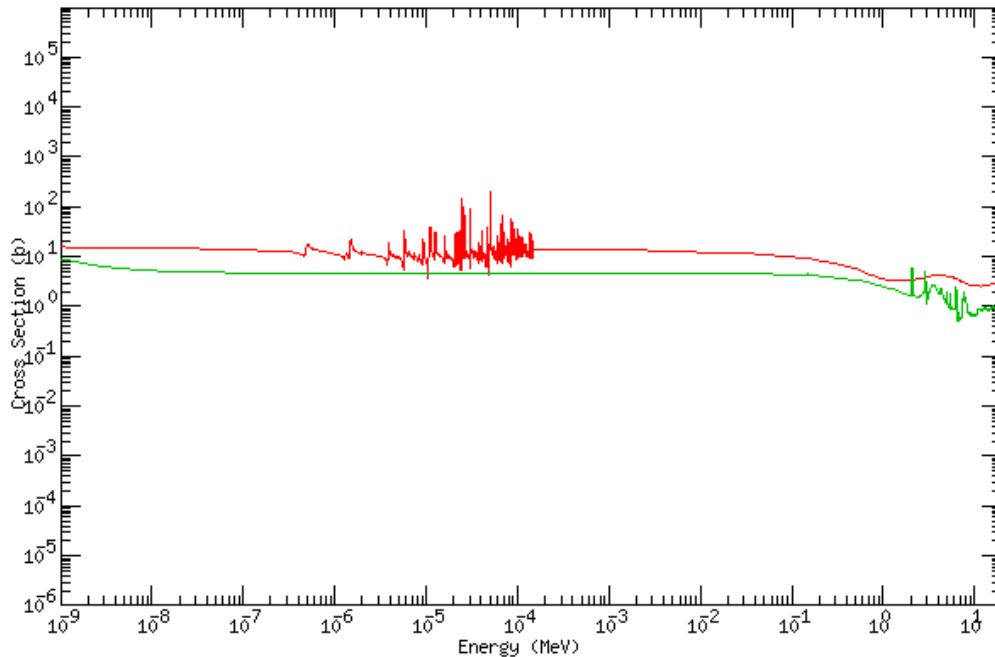


Fig 11.-Elastic Scattering Cross Section (Graphite green and Np-237 red)

Once the ideal position for the Np-237 target was established the next step was to determine the geometric configuration of the neutron source. There are numerous options for nuclear reactor configurations, however because TRIGA reactors were chosen for this study; two of the most common TRIGA core geometries were simulated. The first geometric core configuration was the hexagonal 1 Megawatt TRIGA reactor discussed in the previous paragraphs. The second design was a based on 10 MW TRIGA core fuel [6] rectangular configuration with smaller fuel pins and a smaller core. The dimensions of the rectangular core are 40.8 x 40.8 x 76.5. There are 480 20% U-235 enriched zirconium hydride fuel pins with a 1.3 cm diameter and a 55.9 length (Figure 12). The rectangular smaller pin fuel core was chosen for the study because is the standard shape core for TRIGA reactors above 3 MW. The rectangular core reactor was modeled to determine if and how a different shaped neutron source affects the activation of the Np-237 target. This configuration did not yield the desire results due to the fact that having a smaller core means that a thicker target is needed. A thicker target signifies that less material will get activated due to material attenuation and self shielding. Also the smaller pins mean that there is less moderator material inside the fuel yielding a harder neutron spectrum at the outside boundary of the core.

After the first two models configuration simulations were completed and the results were analyzed it was concluded that the hexagonal shape core model was better suited for Pu-238 production. It was also concluded that instead of using power as a design constraint, average neutron flux at the Np-237 target should be the determining design parameter of the final nuclear reactor configuration. In addition it was also established that in order to optimize and to have a more uniform target activation a reflector material or a uranium blanket should be place surrounding the Np-237 target to minimize neutron leakage. From the conclusions draw from the first two nuclear reactor configuration a third nuclear reactor model was created.

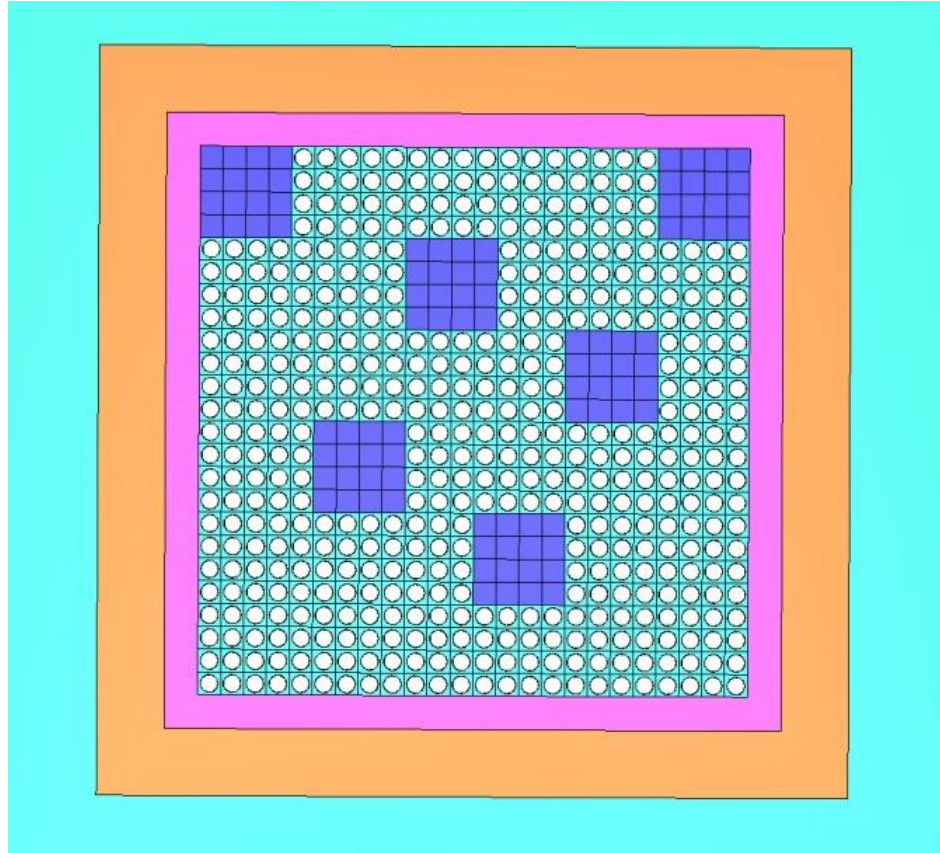


Figure 12.- Cross Section of the Hexagonal Nuclear Reactor Core

The third and final nuclear reactor configuration or Annular Flowing Target TRIGA Reactor (AFTTR) was based on the hexagonal 1MW TRIGA modeled in the first stage of the study. The AFTTR has a larger hexagonal core than the 1 MW TRIGA with more fuel element rows (Figure 13) . The zirconium hydride fuel pins contained in the AFTTR model are also the same size as the 1 MW TRIGA fuel elements. Three different configurations of the AFTTR model were simulated. The three configurations have the same core design dimension; the models differ in the materials used to surround the Np-237 target. The modifications were made to minimize neutron leakage and to increase the average flux at the target. The main purpose of AFTTR model was to establish the amount of flux necessary to produce desire Pu-238 production and to have a preliminary design of a reactor that can produced 1.5 Kg of Pu-238.

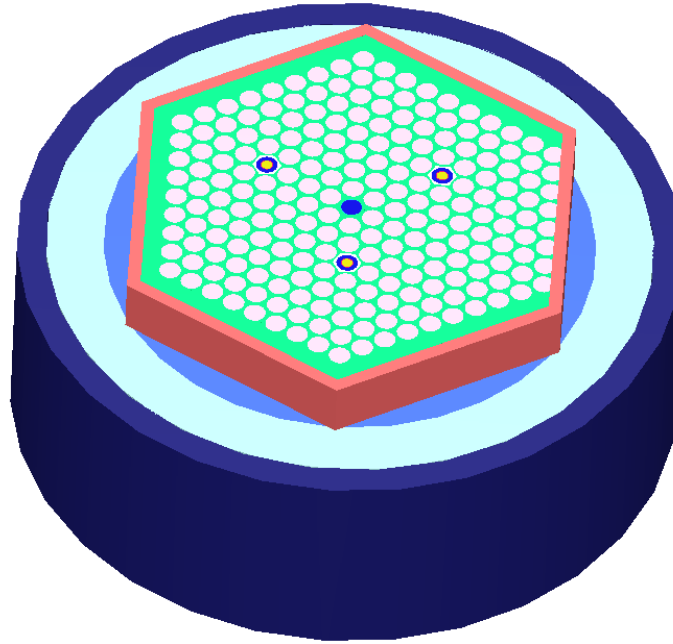


Figure 13.-Final AFTTR configuration

The production results of the final configuration with different reflector blankets are presented in Table 2. The results show how given the same neutron source strength the better results are achieved using the uranium blanket reflector configuration, however the desired production of 1.5 kilograms of Pu-238 is achieved with the graphite reflector.

Table 2.- AFTTR Configuration Results

	Uranium	Graphite	Water
<b>Thermal Flux n/(cm<sup>2</sup>*sec)</b>	2.80E+13	2.43E+13	1.78E+13
<b>Total Flux n/(cm<sup>2</sup>*sec)</b>	7.34E+13	5.99E+13	4.28E+13
<b>Pu-238 kg/yr</b>	1.71	1.51	1.05

### Reactor Design Summary

The feasibility nuclear reactor model study was able to determine three key design parameters

- The position of the Np-237 target in the nuclear reactor configuration
- The geometric configuration of the nuclear reactor
- The average amount of neutron flux target needed for production of 1.5 Kg of Pu-238 per year

The study concluded that having the Np-237 target outside the reactor fuel boundaries does not decrease criticality, therefore is not a limiting factor in the production of Pu-238. The study also established that the reactor configuration for producing Pu-238 has to be a hexagonal core with a

circular Np-237 target surrounded by a reflector blanket. In addition it was determined that the desired production is achieved with a configuration that includes a graphite reflector blanket surrounding the Np-237 target. The feasibility study preliminary dimensions of the core along with fuel element dimensions, size and thickness of the target and D<sub>2</sub>O gap thickness were determined and are shown in Table 3.

Finally the study established that it is possible to produce enough Pu-238 using a liquid target outside the reactor core if the average neutron flux required at the target is achieved. A total average neutron flux of  $5.99\text{E}+13 \text{ n/cm}^2\cdot\text{sec}$  at the Np-237 target is needed produce 1.5 Kg of Pu-238. This is the first time that a study using this configuration with an encapsulated target have proven to be effective. The production can be further maximized by having a fuel blanket surrounding the Np-237 target however this configuration will increase design and production cost. Further studies should be perform to determine different methods to maximize the flux spectrum available at the target in order to decrease the target mass and increase production. The feasibility study presented in this section shows that production can be achieve using a TRIGA reactor however any reactor that satisfies the target placement, core configuration and flux requirements could be used to produce Pu-238 in a encapsulated continuous manner.

Table 3.-AFTTR Core Dimensions

<b>Core Diameter (cm)</b>	70.46
<b>Core Height (cm)</b>	58.4
<b>Fuel Element diameter (cm)</b>	3.64
<b>Fuel Element Height (cm)</b>	38.2
<b>Number of Control Rods</b>	3
<b>Enrichment of the fuel</b>	20%
<b>Np-237 Thickness</b>	5
<b>Np-237 Height</b>	38.2
<b>D<sub>2</sub>O Gap Thickness</b>	5
<b>D<sub>2</sub>O Gap Height</b>	38.2
<b>Graphite Reflector Target Diameter (cm)</b>	5
<b>Graphite Reflector Target Height(cm)</b>	38.2

### III.c. System modeling

To establish various correlations with reactor power and irradiation time, a parametric trade study has been performed. In essence, the flow of capsules was modeled around a core whose diameter varied as the square root of the power level. The diameter dictated the length of the pipe surrounding the core which in turn dictated the capsule velocity in order to match the desired irradiation time. Given the capsule velocity, the length of pipe necessary for the decay

time to elapse was determined. The pipe lengths dictated the mass of Np-237 residing in the reactor tank. Thus, by using the estimated production rate of Pu-238 per kg of Np-237 versus irradiation time and the maximum concentration of Np-237 per capsule, we can parametrically estimate the annual Pu-238 production as a function of neutron flux level.

The trade study used a TRIGA as a default reactor design. Possibly, other vendors may have reactors available in the 1 to 10 MW power class but the TRIGA is a well known design and is a good example of the class. In addition, we had measured neutron spectra available for the 1 MW TRIGA system. The actual correlation between neutron flux level and power level may not be well established for all different reactor types. We assumed that the flux in the same position in a TRIGA reactor is linear with power level. Thus, we assumed a  $1.4 \times 10^{13}$  n/cm<sup>2</sup>-s per MW for the trade study.

Results for a single point design for a 5 MW reactor are summarized in Table 4.

Table 4. Point design of the Pu-238 production system

Annual Production Pu-238(kg/yr)	2.25
Capsule diameter (cm)	4.0
Capsule length (cm)	4.0
Neptunium concentration (gm/cm <sup>3</sup> )	0.432
Feedline diameter (cm)	6.0
Reactor height (cm)	38.0
Reactor diameter (cm)	70.0
Irradiation time (d)	18.
Decay time (d)	12.
Processing time per capsule (s)	4392.
Capsule velocity (m/hr)	0.033
Number coils core	6.3
Number coils decay line	0.7
Length feedline core (m)	14.0
Length decay line (m)	9.4
Mass Np around core (kg)	7.65
Mass Np in decay line (kg)	5.1

The analysis allows for variable capsule sizes, variable reactor power and neutron flux levels, and determines the mass of Np-237 and the amount of Pu-238 produced. In short, at a given reactor power level, there is an optimum amount of Pu-238 that can be produced due to the requirement of a fixed capsule velocity around the core. Consequently, the trade study does not vary the cycle time of the capsules to just produce the 1.5 kg/yr but, instead, determines the mass



of Pu-238 produced by that power reactor for a cycle time commensurate with the capsule velocity through the reactor.

The results in Table 4 show that for roughly 13 kgs of Np-237 in the entire line, we can produce the 2.25 kg/yr of Pu-238 in a 5 MW reactor. This value varies with reactor power. The amount of Np used is an important factor both from the cost standpoint, the latest quote is \$200,000/kg Np-237, and from a waste production standpoint, i.e. more fissions occurring in the Np.

The optimum amount of Pu-238 produced as a function of reactor power is shown in Figure 14. The Figure shows a slightly non-linear dependence with improved performance at higher powers. This dependence actually reduces the price per kg of the Pu-238 (see cost section below).

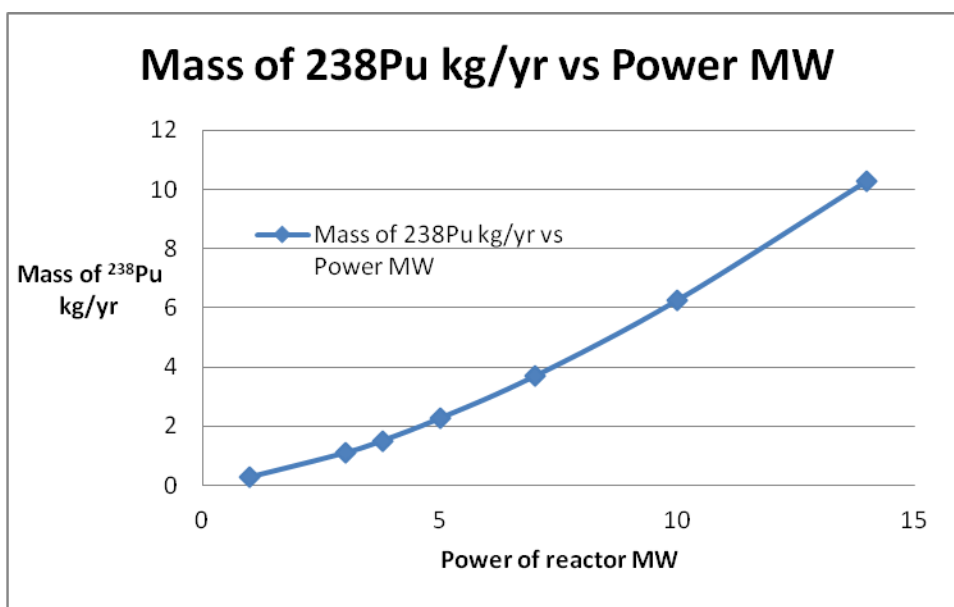


Figure 14. Optimum mass of Pu-238 produced as a function of reactor power. The neutron flux used in the optimization is assumed to be linear with power.

The trade study also allowed confirmation of the optimum irradiation time. One possible Figure of Merit (FOM) is the mass of Pu-238 produced per unit mass of Np-237 per time of irradiation. The desired goal is to have the most Pu produced for the least amount of Np in the shortest time. The results of the trade study were used to generate the FOM shown in Figure 15. Even though the calculation of Pu-239 production shown in Figure 8 indicated a peak irradiation time of near 40 days, Figure 15 clearly shows that an optimum time is in the 12-15 day interval. By adding in the factor of mass Pu-238 versus mass fission products, which should be maximized also, the optimum time of 18 days was found for the irradiation time.

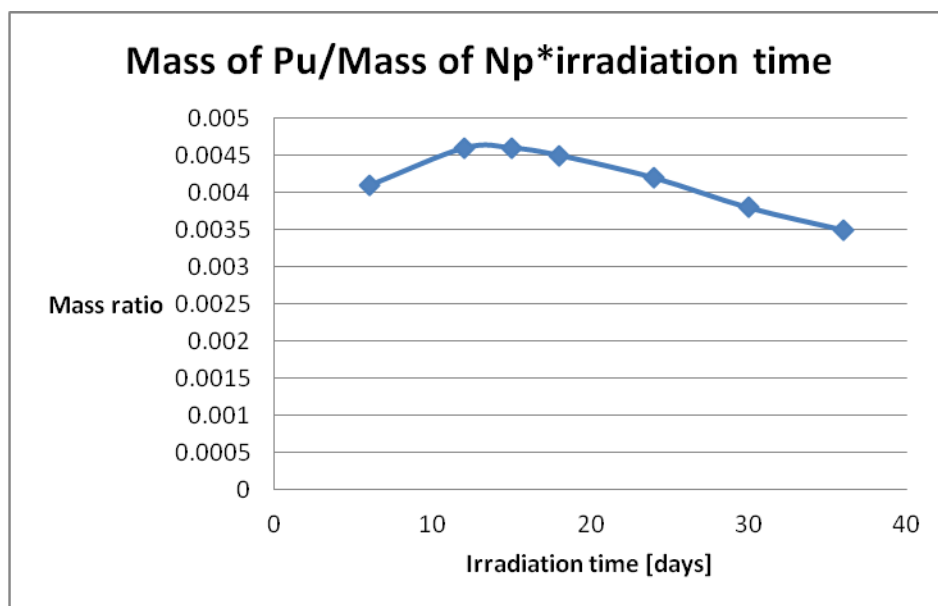


Figure 15. Plot of the Figure of Merit versus irradiation time indicating an optimum near 12 to 15 days.

#### III.d. Chemical Separation

To demonstrate the chemical process for isotope separation is valid and provides the product yield needed for  $^{238}\text{Pu}$  production the following work is proposed to accomplish this.

It is proposed to produce Pu-238 from Np-237 in a nuclear reactor where the reactor is fed with capsules of an acidic aqueous solution of Np-237. A small amount of the Np-237 is transformed into Pu-238 after thermal neutron exposure followed by radioactive decay in a cooling pool. The purpose of this proposed experimental work is to validate that a chemical separation can be performed on the effluent of the nuclear reactor for the mix of isotopes in solution. This experimental program envisions a separation assuming a 4 to 6 day irradiation time. As a result, it is necessary to separate the quantity of Pu-238 from the Np-237 solution. Both of these actinides are in the +4 (IV) valence state for this experiment. A demonstration of the critical separation will be performed as part of this work. To allow this experiment to be performed within the NRC license for University of Utah a lower concentration of Np-237 and Pu-238 will be used for the crucial separation experiment. This experiment, should it be successful, will clearly demonstrate that this type of separation is technically feasible.

## Review of Separation Technology

Historically the PUREX process consisting of liquid-liquid decantation has been used in the US but this separation technology has been superseded by ion exchange due to generating less radioactive waste.

The envisioned chemical separation is a challenging problem that can be performed by ion exchange using an anionic resin like Dowex resin. The common procedure consists of the adjustment of neptunium ion at Np (IV) and the adsorption of Np (IV) nitrate complex, i.e.  $\text{Np}(\text{NO}_3)_6^{-2}$ , on the anion-exchange resin from 7 to 8 M  $\text{HNO}_3$  solution. The anionic nitrate complexes of Pu (IV). Pu (IV) is eluted as Pu (III) with a mixture of 6 M  $\text{HNO}_3$  + 0.05 M Fe(II) sulfamate + 0.05 M hydrazine. Np (IV) is then recovered by elution with 0.3 M  $\text{HNO}_3$ . Maiti et al. [7] developed a method for the sequential separation of actinides by anion ion exchange. Np (IV) and Pu (IV) in 9 M  $\text{HCl}$ –0.05 M  $\text{HNO}_3$  solution are adsorbed on the anion ion exchange resin. Pu (IV) and Np (IV) are eluted successively using 9 M  $\text{HCl}$  and 0.05 M  $\text{NH}_4\text{I}$  and 4 M  $\text{HCl}$  and 0.1 M  $\text{HF}$ , respectively. Finally there is a group of ion exchange resins referred to as chelating resins [8], e.g. TEVA, TBP-loaded Amberlite XAD-4 resin and Diphonix, that are useful for this separation.

The separation process as it is to be practiced in this work consists of a modified process using key attributes of the above referenced process. It will include: 1) the Dowex anion ion exchange resin must first be conditioned with concentrated Nitric acid to be useful for this separation process, 2) the ion exchange resin is used then to treat the mixed nitrate solution where only Np (IV) and Pu (IV) are extracted from the 7-8 Molar (M) nitric acid ( $\text{HNO}_3$ ) solution and 3) the ion exchange resin with actinides loaded is then treated to remove Pu(IV) which is eluted as Pu (III) using 2 bed volumes of stripping solution consisting of a mixture of 6 M  $\text{HNO}_3$ , 0.05 M Fe(II) sulfamate and 0.05 M hydrazine. 4) Should thorium be present Th (IV) is eluted with 2 bed volumes of 8 M  $\text{HCl}$ . And 5) Np (IV) is then recovered by elution with 2 bed volumes of 0.3 M  $\text{HNO}_3$ . The resulting Np (IV) nitrate solution is mixed with nitric acid resulting from step 2 to bring it to 7-8 M in nitric acid and passed through the nuclear reactor again to facilitate the production of more Pu-238.

In this process the ion exchange medium is reused over and over in the process using two beds one undergoing adsorption and the other undergoing stripping. Often another is also purchased to be on standby for the adsorption process should there be any problems. The key to the separation is the adjustment of the valence of the Pu (IV) produced by nuclear reactor to Pu (III). This is done during elution/stripping by the addition of Fe(II) sulfamate and hydrazine during step 3. There are several commercial examples of this ion exchange separation with separation efficiencies of 99.9% [8] Water and urine samples with even lower Np concentrations in the low parts per million have shown separation efficiencies of ~96% [9,10].

The research work to be done prior to the separation experiments will develop a model of the ion exchange separation process with all stripping solutions. This work will provide an understanding of what to expect with the experiments. Crucial separation experiments of this type will be performed with solutions with smaller concentrations of Np and Pu based up the ability of the University of Utah to obtain a license from the NRC for these experiments. An initial

assessment of the potential for radiation exposure of a solution containing 10 mg Np-237 and 10 mg Pu-238 is ~1 mCi. Analysis of all the resulting solutions for their content of Np and Pu will be measured by KSU and the data will be used to improve the model of ion exchange separation process so that it can be extrapolated to higher concentrations typical of continuous operations. In addition the amount of Np-237 and Pu-238 held on the ion exchange resin after the adsorption/strip cycle will also be determined by KSU for an assessment of potential radiological waste streams.

This experiment will provide a definitive proof of concept for the separation process envisioned for this new process to produce 1.5 Kg Pu-238 for deep space power sources.

#### **IV. Experimental studies**

##### **IV.a. Sphere fabrication**

Early in the Phase I study, the issue what form the final product from the irradiation should take was raised. The issue hinges on the current process to make clad pellets for MMRTGs or ASRGs. The process involves ball milling of the Pu-238 powder and then sintering the ball milled product into shape. During the ball milling stage, sub-micron particles are produced which are very mobile and difficult to contain. All human exposures to Pu-238 at LANL over the past few decades are the result of sub-micron particles migrating through the seals on glove boxes.

The quantized, continuous flow process produces resin spheres covered in the Pu-238. By washing these spheres with a suitable solution, the Pu-238 is removed and collected as a solution. Conceptually, this solution can be fed directly into a process that creates large diameter, perhaps 100 micron, spheres. These spheres would not be mobile not be able to breach glove box seals. Consequently, the “back end” process was seen as an extension of the goal of the Phase I project to reduce costs of making radioisotope power units. An experimental effort was developed to demonstrate the production of spheres directly from an acidic solution containing a trans-uranic element.

Collaborators at the University of Michigan’s Neutron Science Laboratory have established a sol-gel microsphere production rig suitable for work with radioactive materials. The sol-gel apparatus uses the internal gelation method to fabricate spheres with tunable diameters less than a millimeter without dust generation. During the development and construction of the sol-gel rig, a non-radioactive cerium surrogate for plutonium was used successfully. Currently, depleted uranium is being acquired for use in the system to act as a nearer surrogate for plutonium.

Work with surrogates has indicated that the internal gelation sol-gel fabrication technique will offer substantial benefits over current precipitation and powder processing methods for Pu-238 sources. Current methods involve an oxalate precipitation of plutonium that yields a powder morphology requiring ball milling, which results in respirable fines. A major advantage of the sol-gel method is that no powders are produced. Plutonium nitrate obtained directly from separation of neptunium following reactor production is used as the feed solution for the sol-gel process. Plutonium remains in solution until it is formed into microspheres of the prescribed size.

These gelled microspheres can then be washed, sintered, and mixed with tungsten powder for spark plasma sintering into the final fuel form.

Eventually, depleted uranium spheres will be used as a surrogate for Pu spheres. The spheres will be sintered using the Spark Plasma Sintering (SPS) furnace at the Idaho National Laboratory. The SPS has been used by the CSNR to fabricate a variety of shapes and materials with variable porosity and density profiles. The goal is to produce a sintered pellet that is identical to the currently produced pellet for the General Purpose Heat Sources (GPHS) used in the MMRTG and ASRG.

#### IV.b. Irradiation validation

Although the computational models used in the Phase I effort are considered “benchmarked” and use the most up-to-date cross section sets, the issue of accuracy of the estimated production is still valid. As seen earlier in Figure 1, the absorption cross section of Np-237 has a resonance region for neutrons with energies between 1 eV and 100 eV. This region is difficult to calculate accurately. Consequently, the Phase I effort sought to perform a validation experiment early on.

After contacting several university based research reactors around the country, the Kansas State University (KSU) reactor was chosen to perform the irradiation. Primarily, the choice hinged on the ability to run the reactor continuously for 4 days. Other facilities would only run during the day shift which would have made analysis of the production rate very complicated.

Original discussions intended to perform the irradiation around January, 2012. Preparations were started to send a 10 gm sample from the INL storage to KSU. Unfortunately, the INL experienced a radiation exposure in late November, 2011 that forced the closure of the vault that contained the Np sample. Subsequent efforts to purchase a sample from the Oak Ridge National Laboratory were unsuccessful.

The vault at INL was finally accessed in July, 2012. Efforts are underway to get the Np sample to KSU. However, KSU cannot easily perform the 4 day irradiation in the school term. Consequently, the irradiation is currently planned for late December. The Phase I contract has been granted a no-fee extension which will allow an addendum to this final report to be submitted at the end of the calendar year.

### V. Cost Estimates

The current method of production by the DOE has major costs associated with handling large quantities of highly radioactive material at a given time. This necessitates large facilities, hot cells with remote manipulation, and transportation costs. The target pins have to be stored in a shielded facility for the required decay time to convert the Np-238 to Pu-238. They are then moved to the area where they have to be dissolved in the nitric acid solution prior to flowing into the extensive separation lines. In short, the facility footprints and the staff associated with the complex handling scheme are significant costs of the current process.

Qualitatively, the continuous feed method does not require the facility space to decay the irradiated product because the feedline is designed to allow the decay within the water tank of the reactor, i.e. the velocity of the capsules matches the required irradiation time in the core as well as the decay time through the tank. Because we are treating one capsule at a time, the separation lines are very modest. Early indications are that the entire separation system may be able to sit on the top of the reactor or immediately nearby so that a separate facility is not required. The cost estimate for the separation system is shown in Appendix A.

Initially, the Phase I project started evaluating two possible scenarios for locating the facility: 1) a private entity builds a new facility on a “green field” site and transports the Pu-238 product to a DOE laboratory for fabrication, or 2) a private entity leases land from the government at a DOE Laboratory. The advantage of scenario 1 was that the land may be cheaper and it would be owned by the private entity which would be attractive to investors. The disadvantages were that all security costs and general infrastructure must be created fresh and would be part of the overhead costs. In addition, the transportation costs over public lands from the reactor facility to the DOE national laboratories may be significant. The advantages of scenario 2 were that the security and utilities infrastructure exists at the DOE sites as well as safety and handling procedures. Also, the transportation would only be on-site and never cross public land. The disadvantages are an increased cost of the land and operation on government controlled land.

Early discussions with personnel at the INL allowed the project to quickly determine that scenario 1 was not a competitive concept. In essence, the amount of Np-237 that would be present on site will make the facility Category II. This requires significant physical security to be in place. In addition, the costs of shipping the Pu-238 from the reactor to a DOE site for fabrication would also require significant expense, especially if done on a weekly basis. Finally, Scenario I would also entail licensing by the Nuclear Regulatory Commission (NRC) which would entail longer approval periods and higher costs.

Scenario II entails placing the facility at an existing DOE Laboratory site that already has Category II facilities. In this way, physical security and infrastructure are present and the human security can be shared with the other facilities. The DOE has several national laboratories around the country but many of them are now surrounded by residential housing or otherwise unsuitable to house a new reactor. Among the DOE labs that could support a reactor facility that produces Pu-238, the costs of human security can vary significantly.

The INL has the lowest costs associated with its facilities in the DOE complex. Consequently, the Phase I recommendation is for the production facility to be placed at the Materials and Fuels Complex (MFC) at the INL. A preliminary cost estimate for sharing the human security to cover the Pu production facility is estimated to be \$255,000 fully burdened. This recommendation has the added benefit of reducing the issues of transporting any Pu-238 as the Space and Security Power Systems Facility (SSPSF) which assembles the MMRTGs is in the MFC site. Thus, the production, processing, and assembly could be accomplished in one location.

## V.a. 5 MW Point Design

The design of the separation system is shown in Appendix A. The total cost estimate methodology, cost of separation system, and total capital costs are shown in Appendix B. These Appendices were made for a point design of a 5 MW reactor. The ROI was based on an assumed price for the Pu-238 to be \$6M.

Using the Guthrie and Lang methods the bare module cost are used to determine other costs needed to calculate the total capital investment. These capital cost categories include the cost for utility plants including, electricity, water and steam plant for water distillation, site preparation costs, service facilities, contingencies and contractor fees, cost of land and plant startup. Working capital is estimated at 17.6% of total capital investment since the Np-237 and Pu-238 inventory must known to calculate this figure more accurately. As a result of this work the total capital investment is \$66 million. Given the cost estimation methods used this value is  $\pm 35\%$  assuming all major chemical separation units have been accounted for. The largest cost associated with the total capital investment is working capital accounting for more than 50% of the total capital investment. It should be noted that the site costs associated with an extension of an existing security fence is included in the site preparation costs associated with the total capital investment.

Table 5. Total Capital Investment

Allocated Cost for Utility		
Plants	\$	101,058
Site Preparation Costs	\$	1,614,422
Service Facilities	\$	2,152,563
Contingencies & Contractor		
fees	\$	1,937,307
Cost of Land	\$	331,363
Cost of Plant Startup	\$	1,658,117
Working Capital	\$	22,521,417
Total Bare Module Costs	\$	35,762,816
Total Capital Investment	\$	66,079,064

## Operating Expenses - Separations

From the prices of raw materials including \$200,000 per kg for Np-238, the cost of capital at 6.5% per year, cost of labor and waste disposal costs provide all of the annualized costs to produce Pu-238 on a per kg basis. As a result, the cost to separate Pu-238 in this process is \$3,245,104 per kg Pu-238. Labor costs in total account for \$1.26 million/yr half of which is required for security and half for nuclear/chemical operators. Given that it is estimated that the sale price is \$6,000,000 per kg, this process will be economically viable should the other expenses including the cost of operating the nuclear reactor and electroplating be less than \$2,000,000/yr as is likely the case.

Table 6 Operating Expenses - Separations

		Operational Expenses
		\$ per kg Pu-238
Cost of Capital @	6.50%	\$ 1,780,093
Resin costs		\$ 2,862
Np-237		\$ 200,000
Steam		\$ 0
Electricity		\$ 3
Cooling Water		\$ 91
Process Water		\$ 16
Liquid Waste		\$ 581
Labor Cost		\$ 633,600
Security		\$ 630,720
Process Costs	Total	\$ 3,245,104 \$ per kg Pu-238
		+/- 50% estimate
Sale Price		\$ 6,000,000 \$ per kg Pu-238

#### V.b. Cost Trade Study

Preliminary results of the cost estimate are very dependent upon the reactor power required. The initial capital cost depends almost linearly on the reactor power level and is the largest single component. In addition, the amount of Np-237 present in the reactor is dependent on the power level. Finally, due to the requirement to fix the capsule velocity for the appropriate irradiation time, the optimum amount of Pu-238 produced, thus the revenue stream, varies with reactor power. Figure 16 shows the mass of Pu-238 produced versus reactor power. (To be clear, the power level shown is a correlation between neutron flux and power level. The measured flux of  $1.4 \times 10^{13}$  for a 1 MW reactor is linearly scaled in the figure, i.e. a 5 MW reactor will have  $7 \times 10^{13}$  n/cm<sup>2</sup>/s.) Figure 16 also shows the price per kg of Pu-238 to be charged to the government in order to make a) a 20% ROI and b) a 0% ROI, i.e. if the government elected to use this process and not make a profit. The figure shows that a 5 MW reactor could produce 2.25 kg of Pu-238 per year and would need to sell material for \$7.8 M/kg for a 20% ROI and \$3.5 M per kg for a 0% ROI. For a 10 MW reactor, the prices are \$4.3 M and \$ 1.6 M for a 20% and 0% ROIs respectively. This is because the 10 MW system will optimally make 6.25 kg/yr. Thus, even though the initial cost for the reactor goes up, the revenue goes up faster and the cost per kg is less.



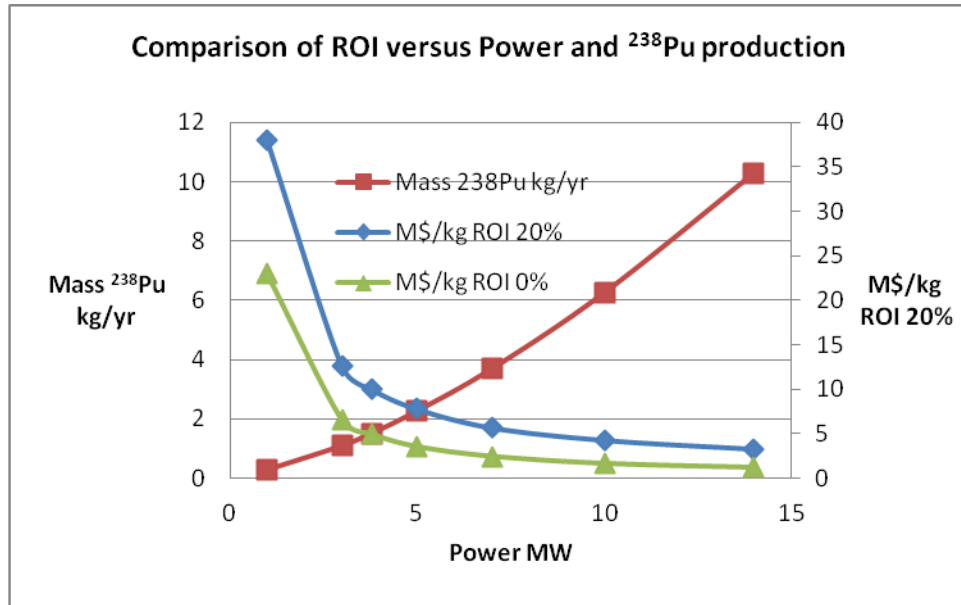


Figure 16. Plots of Pu-238 mass produced, price per kg Pu for 20% ROI and price per kg PU for a 0% ROI versus reactor power.

## VI. Future Work

The main product of the Phase I effort has been to show computationally that a low power reactor can produce a high grad product of Pu-238 in the amounts specified to support the NASA mission plan. One of the main questions remaining is whether or not a mechanical system can be built that efficiently moves the required amount of target material down around the reactor core and up to the top of the reactor. In addition, the heat generated in the capsules may restrict the concentration of Np-237 allowed in the capsule. The heat generated is dependent upon the flux level, the concentration, and the heat transfer through the walls of the capsule to the surrounding water flow. On other aspect that needs to be demonstrated is the separation of the irradiated solution into respective components, i.e. Plutonium (product), Neptunium (recycled), fission products (waste), and nitric acid solution (recycled). Finally, the ability to fabricate a sintered pellet using large diameter spheres with the correct porosity and density profiles as that produced for MMRTGs is needed.

### VI.a. Piping and Mechanical Needs for Capsule Movement

The key piece of the process is the mechanical aspects of the piping and flow of the capsules. This flow system can only be modeled crudely and it must be built to demonstrate that it will perform properly in a typical nuclear reactor environment, i.e. a water tank. The system of piping needs to be tested in a standard water tank to show how the capsules will behave in the pipe and to determine the actual mechanical nature of the system.

Some of the key mechanical features of the system that need to be determined are: the pressures needed to pump the capsules at the flow conditions which correspond to the residence time needed for  $^{238}\text{Pu}$  production, the length of the pipe for the process and the behavior of the capsules inside the pipe line. The capsules also need to be tested for buoyancy which is needed to demonstrate the safety of the capsules in case of a line break. The heat transfer analysis will also aid in the determination of the materials for the pipe and capsule.

#### VI.b. Heat Transfer

Simulate the heat transfer in the product line and nuclear reactor to determine the operating temperatures and to determine the best material for the capsules. This is necessary because of the heating from the fission rate in the product line and the heating rate of the nuclear reactor.

To determine the heat transfer for the project, multiphysics modeling software is best suited for the task to simulate the flow of coolant (CFD) and heat transfer in the system. The simulation will quantify the heat flux, temperature gradients in the product line, capsule, nuclear fuel, the needed convective heat transfer coefficient ( $h$ ) from the coolant for the fuel and the product line. This simulation will allow a determination of the best material to handle the temperature gradients in the product line and the capsule.

### VII. Conclusions

In the Phase I NIAC grant, the CSNR has evaluated the feasibility of using a low power, commercially available nuclear reactor to produce 1.5 kg of Pu-238 per year. The impact on the neutronics of the reactor have been assessed, the amount of Neptunium target material estimated, and the production rates calculated. In addition, the size of the post-irradiation processing facility has been established. Finally, as the study progressed, a new method for fabricating the Pu-238 product into the form used for power sources has been identified to reduce the cost of the final product. In short, the concept appears to be viable, can produce the amount of Pu-238 needed to support the NASA missions, can be available within a few years, and will cost significantly less than the current DOE program.

The alternative method to produce Pu-238 is to continuously flow an encapsulated aqueous solution containing a high concentration of dissolved Np-237 in a water carrier stream. Once the optimum irradiation period is completed, the encapsulated target slowly moves through the reactor's water tank which allows for time for the decay of Np-238 to Pu-238. This process allows small quantities of Pu-238 to be processed on a weekly basis so that a much smaller, and less costly, facility is needed to accumulate the Pu-238. One other aspect that has come out of the Phase I effort is the recognition that the new process will produce a substantially smaller waste stream of radioactive acidic solution, mixed waste, which has to be stored or processed. Thus, the method will produce substantially reduced costs.

In addition to the technical assessment of the production, the study sought to determine the answers to two major issues: 1) given a sufficient price per kg of Pu-238, could a sufficient return on investment (ROI) be possible so that private venture would pay the up-front capital

costs saving the government this requirement in times of diminished budgets, and 2) is it more cost effective to install the new reactor on private land with private operations rather than locating the reactor at a DOE facility? The results of the study indicate that a 20% ROI is possible if the price per kg paid for the material is commensurate with the last known, circa 2007, asking price from Russia. The results also show that, due to lower security and transportation costs, the only responsible option is to locate the reactor at the Idaho National Laboratory site of the DOE.

The Phase I effort utilized experimentally measured neutron spectra from a 1 MW TRIGA reactor at the Kansas State University to estimate the Pu-238 production. Although several reactor types may be available for Pu-238 production, the TRIGA was used to model the production rates due to the large database regarding neutron flux and costs. By assuming a linear scaling of the neutron flux but keeping the neutron spectra the same, the production rate of the Pu-238, the concentration of Pu-236, and the amount of fission products could be calculated. The calculations show that an irradiation time of between 15 to 18 days with a 12 day decay time is optimum. Pu-236 contamination should be less than 2 ppm. The amount of fission products is estimated to be 150 gms/yr.

Using an 18 day irradiation time, the production rate versus neutron flux, i.e. reactor power, was determined. The trade studies indicate that a reactor at 3.8 or 10 MW can produce 1.5 or 6.25 kgs/yr of Pu-238 respectively. If a 20% return on investment is required, i.e. if the facility is privately funded, the price of the Pu-238 sold to the DOE would have to be 10 \$M/kg and 4.3 \$M/kg respectively. If no ROI is required, i.e. the US government funds the facility, then the price is 4.9 \$M/kg and 1.6 \$M/kg for the 1.5 kg and the 6.25 kg respectively. In either case, the results of this study indicate that the 1.5 kgs/yr of Pu-238 can be produced in a new facility within a 3-4 year timeframe for around \$50M and return a 20 % ROI to an investor group.

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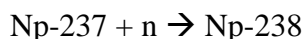
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9. <http://radchem.nevada.edu/classes/rdch710/files/neptunium.pdf>
10. <http://sti.srs.gov/fulltext/ms2002997/ms2002997.html>

## Appendix A -- Chemical Separation Unit Design for Pu-238 Production

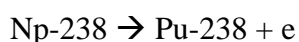
Pu-238 for space power supplies associated with missions to the outer planets in the solar system are in need of Pu-238 supplies that are essentially lacking in the nuclear stockpile. This work provides a new route to produce Pu-238 from Np-237 using capsules of Np solution in a flow circuit that goes into the core then the pool of a nuclear reactor to expose Np-237 to thermal neutrons at high flux followed by cooling off period in the reactor pool. As the material is removed from the nuclear reactor it undergoes a chemical separation process which recycles the untransformed Np and removes the Pu-238 product as a nitrate solution. The capital cost of this project is \$35.7 million with \$35.2 million for the nuclear reactor and the security fence extension needed to encase it in an existing CAT.-1 security envelope at INL. The costs for this process on a per kg of Pu-238 are \$3.2 million ( $\pm 50\%$ ) which is substantially above the sale price of \$6,000,000 giving a return on investment of 4%/yr and a pay-back-period of 5.3 yr. The capital and operating costs are substantially reduced if the nuclear reactor and its security fence can be eliminated giving a processing cost of \$0.56 million per kg Pu-238, a return on investment of 140%/yr and a pay-back-period of 0.3 yr.

### Introduction

It is proposed to produce Pu-238 from Np-237 in a nuclear reactor where the reactor is fed with capsules of an acidic aqueous solution of Np-237. A small amount of the Np-237 (see Table 1) is transformed into Pu-238 after thermal neutron exposure followed by pool cooling according to the nuclear reactions:



in Nuclear Reactor



in Reactor Pool

The purpose of this work is to develop a cost estimate for the chemical separation process required to remove the small amount of Pu-238 from the Np-237 nitrate solution after removal from the capsules. This chemical separation process is critical to the success of this Pu-238 production route.

Table 1 Amount Np-237 Transformed to Pu-238 in a 3 Molal Aqueous Solution in a Nuclear Reactor with a Thermal Neutron Flux of  $4.2 \times 10^{13}$  to  $1.4 \times 10^{14}$  N/cm<sup>2</sup>/s.

Exposure	Pu-238 Produced
18d	0.02 gmPu/gmNp

This economic analysis envisions a chemical separation assuming an 18 day thermal neutron exposure and 12 day pool cooling. As a result, it is necessary to separate a 0.8 % wgt quantity of Pu-238 from a 0.27% wgt Np-237 (metal) solution (3 Molal). Both of these actinides are in the +4 (IV) valence state as they enter and leave the nuclear reactor pool. This work will give the capital and operating costs for this type of separation to determine if it is economically feasible.

## Review of Separation Technology

Historically the PUREX process consisting of liquid-liquid decantation has been used in the US but this separation technology has been superseded by ion exchange due to its generating less radioactive waste.

The envisioned chemical separation is a challenging problem that can be performed by ion exchange using an anionic resin, e.g. Dowex resin. The common procedure consists of the adjustment of neptunium ion at Np (IV) and the adsorption of Np (IV) nitrate complex, i.e.  $\text{Np}(\text{NO}_3)_6^{-2}$ , on the anion-exchange resin from 7 to 8 M  $\text{HNO}_3$  solution. The anionic nitrate complexes of Pu (IV) and Th (IV) are also adsorbed on the resin at the same time. Pu (IV) is eluted as Pu (III) with a mixture of 6 M  $\text{HNO}_3$  + 0.05 M Fe(II) sulfamate + 0.05 M hydrazine. Np (IV) is then recovered by elution with 0.3 M  $\text{HNO}_3$ . Maiti et al.<sup>1</sup> developed a method for the sequential separation of actinides by anion ion exchange. Np (IV) and Pu (IV) in 9 M  $\text{HCl}$ –0.05 M  $\text{HNO}_3$  solution are adsorbed on the anion ion exchange resin. Pu (IV) and Np (IV) are eluted successively using 9 M  $\text{HCl}$  and 0.05 M  $\text{NH}_4\text{I}$  and 4 M  $\text{HCl}$  and 0.1 M  $\text{HF}$ , respectively. Finally there is a group of ion exchange resins referred to as chelating resins<sup>2</sup>, e.g. TEVA, TBP-loaded Amberlite XAD-4 resin and Diphonix, that are useful for this separation.

The separation process as it is to be practiced consists of a modified process using key attributes of the above referenced process; see Figure 5 (substitute). It will include: 1) the Dowex anion ion exchange resin must first be conditioned with concentrated Nitric acid to be useful for this separation process, 2) the ion exchange resin is then used to treat the mixed nitrate solution where Np (IV) and Pu (IV) are extracted from the 7-8 Molar (M) nitric acid ( $\text{HNO}_3$ ) solution and 3) the ion exchange resin with actinides loaded is then treated to remove Pu(IV) which is eluted as Pu (III) using 2 bed volumes of stripping solution consisting of a mixture of 6 M  $\text{HNO}_3$ , 0.05 M Fe(II) sulfamate and 0.05 M hydrazine (strip 1 in Figure 1). 4) should thorium be present, Th (IV) is eluted with other actinides with 2 bed volumes of 8 M  $\text{HCl}$  (this stripping step is not shown in Figure 5 as it only needs to be done on the solution every 100 passes through the nuclear reactor) and 5) Np (IV) is then recovered by elution with 2 bed volumes of 0.3 M  $\text{HNO}_3$ . The resulting Np (IV) nitrate solution is mixed with nitric acid resulting from Position 1 in a recycle solution preparation step shown in Figure 1. After water removal by Reverse Osmosis (RO) used to concentrate the nitric acid stream for recycle to the nuclear reactor to facilitate the production of more Pu-238 and dilute a portion of the nitric acid solution passing through the resin during the first extraction. At the end of all ion exchange operations the ion exchange columns are allow to drain and air dry so that waste solutions are minimized.

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<sup>1</sup> Maiti, T. C., Kaye, J. H., and Kozelisky, A. E. (1992) J. Radioanal. Nucl. Chem., 161, 533–40.

<sup>2</sup> Zenko Yoshida, Stephen G. Johnson, Takaumi Kimura, and John R. Krsul, Neptunium, Chapter 6 in <http://radchem.nevada.edu/classes/rdch710/files/neptunium.pdf>

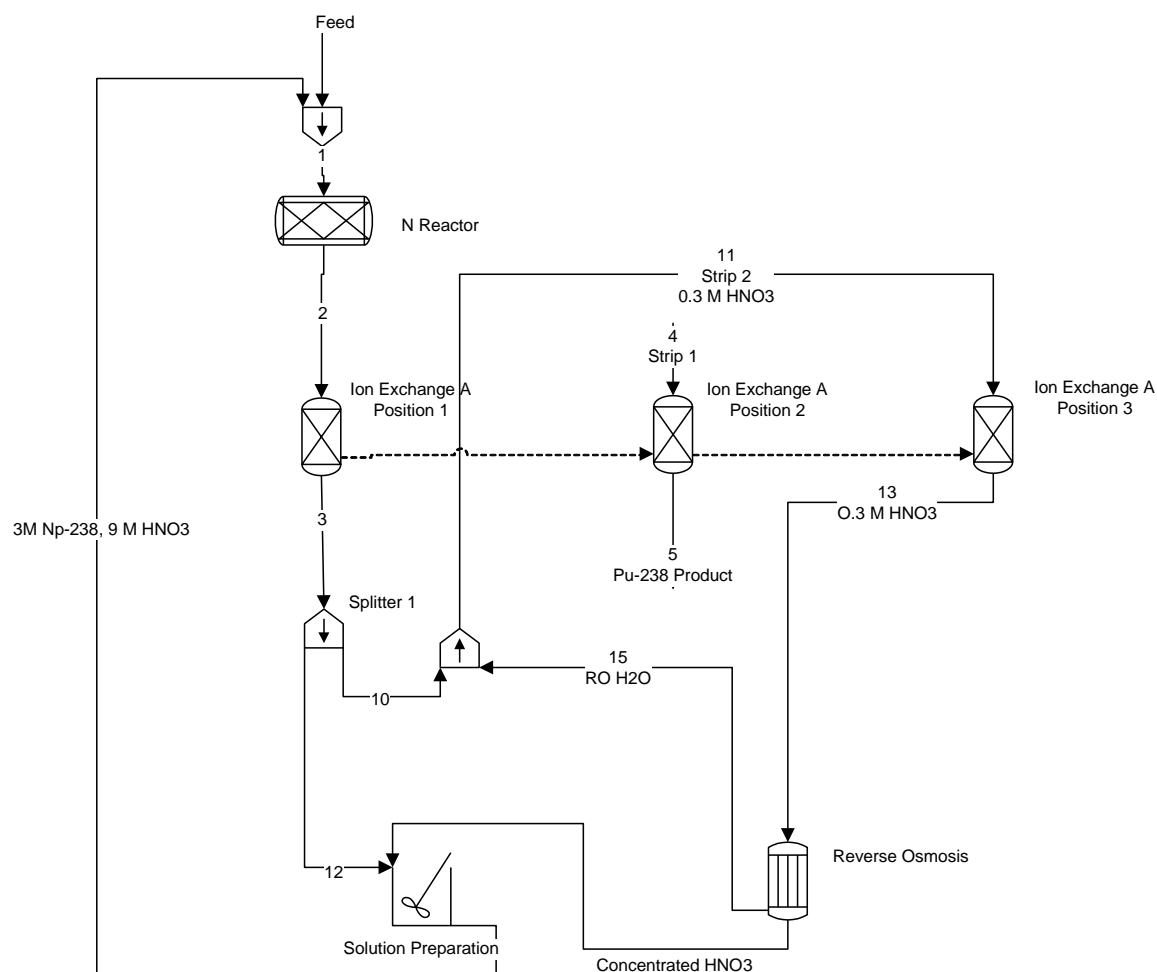


Figure 1 Schematic Diagram of the Pu-238 Chemical Separation Process.

In this process the ion exchange medium is reused over and over in the process using two beds one undergoing adsorption and the other undergoing stripping. Often another is also purchased to be on standby for the adsorption process should there be any problems. The key to the separation is the adjustment of the valence of the Pu (IV) produced by nuclear reactor to Pu (III). This is done during elution/stripping by the addition of Fe(II) sulfamate and hydrazine during step 3. There are several commercial examples of this ion exchange separation with separation efficiencies of 99.9%<sup>2</sup>. Water and urine samples with even lower Np concentrations in the low parts per million range have shown separation efficiencies of ~96%<sup>3,4</sup>.

From this process a solution containing 39% wgt Pu (III) nitrate, 6 M HNO<sub>3</sub> solution (60% wgt), 0.05 M Fe(II) sulfamate and 0.05 M hydrazine is produced. This solution can be separated using a cation exchange column to yield Pu (III) nitrate in nitric acid solution which can be directly electroplated to produce the metal. This final separation and electroplating of Pu-238 are not part of this chemical separation process nor its economic analysis.

<sup>3</sup> <http://radchem.nevada.edu/classes/rdch710/files/neptunium.pdf>

<sup>4</sup> <http://sti.srs.gov/fulltext/ms2002997/ms2002997.html>

## **Mass Balance for Pu Chemical Separation Process**

The mass balance for the process shown in Figure 1 is given in Table 2. Here we see the stream numbers, weight percentages for each of the components, the total flow rate and the individual component flow rates for each stream. In this simulation all extractions are assumed to be 98% efficient while stripping is assumed to be 100% efficient. In addition to this process diagram a steam generator is needed to produce distilled water. The distilled water will be used for the various solutions and also for the water needed to pump the capsules into the nuclear reactor. A coil of pipe will be used for the residence time for thermal neutron exposure and for pool cooling.



Table 2 Mass Balance for the Pu-238 Chemical Separation Process.

	Stream Number	1	2	3	3b	4
	MW	Feed Wgt %	After N Reactor Wgt %	After Extraction Wgt %	In Bed Wgt %	Strip 1 Wgt %
Np(NO <sub>3</sub> ) <sub>4</sub>	357	41.59%	40.76%	1.38%	98.00%	
Pu(NO <sub>3</sub> ) <sub>4</sub>	358	0.00%	0.83%	0.03%	2.00%	
Pu(NO <sub>3</sub> ) <sub>3</sub>	295.9951					
HNO <sub>3</sub>	63.01	19.58%	19.58%	33.04%	0.00%	27.16%
H <sub>2</sub> O	18	38.83%	38.83%	65.55%	0.00%	71.84%
Fe(II) Sulfamate	248.045					0.89%
Hydrazine	32					0.11%
Total		100.0%	100.00%	100.00%	100.0%	100.00%
		kg/hr	kg/hr	kg/hr	kg/hr	kg/hr
Total Flow Rate		0.035	0.035	0.02	0.01	0.000370
Np(NO <sub>3</sub> ) <sub>4</sub>		0.01	0.01	0.00	0.01	
Pu(NO <sub>3</sub> ) <sub>4</sub>		-	0.00029	0.00001	0.00029	
Pu(NO <sub>3</sub> ) <sub>3</sub>						
HNO <sub>3</sub>		0.01	0.01	0.01	0	0.000101
H <sub>2</sub> O		0.01	0.01	0.01	0	0.000266
Fe(II) Sulfamate		-	-	-	0	0.000003
Hydrazine		-	-	-	0	0.0000004

Stream Number		5	5b	7	In Bed	H2O Add
MW	Pu-238 Product	In Bed	Strip 2	Wgt %	Wgt %	Wgt %
		Wgt %	Wgt %			
Np(NO3)4	357		51.25%			
Pu(NO3)4	358		0.00%			
Pu(NO3)3	295.9951	38.95%	0.00%			100%
HNO3	63.01	16.58%	0.90%			
H2O	18	43.86%	47.85%			
Fe(II) Sulfamate	248.045	0.54%	0.00%			
Hydrazine	32	0.07%	0.00%		-	
Total		100.00%	-	100.00%		

	kg/hr	kg	kg/hr	kg/hr	kg/hr
Total Flow Rate	0.000606		0.03		0.01
Np(NO3)4	-	-	0.01	0	
Pu(NO3)4	-	-	0	0	
Pu(NO3)3	0.000236		0		
HNO3	0.000101		0.00		
H2O	0.000266		0.01		0.01
Fe(II) Sulfamate	0.000003		-		
Hydrazine	0.0000004		-		

Stream Number		10	11	12	13	14
MW		SplitExt1 Wgt %	Mix 9+10 For Elution 2	Split Ex1 Prod	After Elution 2 Wgt %	Mix 13+12 Wgt %
				Wgt %		
Np(NO3)4	357	1.38%	0.000771972	1.38%	0.512827032	0.301887541
Pu(NO3)4	358	0.03%	1.57545E-05	0.03%	7.68111E-06	0.000123131
Pu(NO3)3	295.9951	0.00%	0	0.00%	0	0
HNO3	63.01	33.04%	0.018537692	33.04%	0.009038039	0.144882657
H2O	18	65.55%	0.980674582	65.55%	0.478127248	0.553106672
Fe(II) Sulfamate	248.045	0.00%	0	0.00%	0	0
Hydrazine	32	0.00%	0	0.00%	0	0
Total		100.00%	100.00%	100.00%	100.00%	100.00%
				12	13	14
		kg/hr	kg/hr			
Total Flow Rate		0.00	0.01	kg/hr	kg/hr	kg/hr
Np(NO3)4		0.00	0.00	0.02	0.03	0.05
Pu(NO3)4		0.00	0.00	0.00	0.0140	0.01
Pu(NO3)3		-	-	0.00	0.0000	0.00
HNO3		0.00	0.00	-	-	-
H2O		0.00	0.01	0.01	0.0002	0.01
Fe(II) Sulfamate		-	-	0.01	0.0131	0.03
Hydrazine		-	-	-	-	-

	Stream Number	15	16
	MW	RO H2O Wgt %	Np-237 Recycle Wgt %
Np(NO3)4	357		0.411115451
Pu(NO3)4	358		0.000167681
Pu(NO3)3	295.9951		0
HNO3	63.01		0.1973036
H2O	18	100%	0.391413268
Fe(II) Sulfamate	248.045		0
Hydrazine	32		0
Total		100.00%	100.00%

	kg/hr	kg/hr
Total Flow Rate	0.01	0.035
Np(NO3)4		0.01
Pu(NO3)4		0.00
Pu(NO3)3		-
HNO3		0.01
H2O	0.01	0.01
Fe(II) Sulfamate		-
Hydrazine		-

## Appendix B Capital and Operating Cost Methodology

Capital and operating costs for this chemical separation process were determined using industrial standard methods for the estimation of capital and operating costs for the project. Operating costs are determined by accounting for 1) the direct manufacturing costs including feed stocks, utilities including electricity, water (steam, cooling and process water), solid waste treatment, waste water treatment and air-pollution abatement as well as labor and maintenance, 2) operating overhead, and 3) fixed costs including: property taxes and insurance, depreciation, as well as general expenses including: selling (or transfer) expenses, research (direct or allocated) expenses, administrative expenses and management incentives. A mixture of capital costing methods are used for this project including:

1.) Method of Hill<sup>5</sup>

To produce an estimate only two things are needed, a production rate and a flow sheet showing the major pieces of equipment including: gas compressors, reactors and separation equipment. Heat exchangers and pumps are not considered in making the estimate. The estimate uses the Marshall Stevens Process Industry Average Cost Index to account for inflation in this industry. Different types of processes, e.g. fluid vs. solids handling, have different cost estimating factors. Additional factors to account for site preparation, services facilities, utility plants and related facilities can be added. The estimate is accurate to approximately  $\pm 50\%$  and is particularly useful for low-pressure petrochemical plants.

2.) Method of Lang<sup>6, 7, 8</sup> (with improvements by Peters and Timmerhaus<sup>9</sup>)

This method requires a process design, complete with a mass and energy balance and equipment sizing. The estimate uses overall factors that multiply estimates of the delivered cost of all the process equipment including: heat exchangers, pumps as well as gas compressors, reactors and separation equipment. Important factors account for the effects on unit cost of materials of construction, operating pressure and delivery costs of the equipment. The estimate uses the Marshall Stevens Process Industry Average Cost Index to account for inflation in this industry. Different types of processes, e.g. fluid vs solids handling, have different cost estimating factors. Using various Lang factors either the total permanent investment (fixed capital investment) or the total capital investment (including working capital at 17.6% of total permanent investment) can be determined. The estimate is accurate to approximately  $\pm 35\%$ .

3.) Method of Guthrie<sup>10, 11</sup>

The method requires an optimal process design with mass and energy balance, equipment sizing, selection of materials of construction and a process control configuration. To apply the Guthrie

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<sup>5</sup> Hill, R.D., "What Petrochemical Plants Cost," *Petroleum Refiner*, 35(8),106-110, August (1958).

<sup>6</sup> Lang, H.J., "Engineering Approach to Preliminary Cost Estimates," *Chem. Eng.*, 54(9), 130-33, (1947a)

<sup>7</sup> Lang, H.J., "Cost Relationship in Preliminary Cost Estimation," *Chem. Eng.*, 54(10), 117-21, (1947b)

<sup>8</sup> Lang, H.J., "Simplified Approach to Preliminary Cost Estimates," *Chem. Eng.*, 55(6), 112-13, (1948)

<sup>9</sup> Peters, M.S. and Timmerhaus, K.D., *Plant Design and Economics for Chemical Engineers*, 2<sup>nd</sup> Ed., McGraw-Hill, New York, (1968)

<sup>10</sup> Guthrie, K.M., "Data and Techniques for Preliminary Capital Cost Estimating," *Chem. Eng.* 76, 114-42, March 24 (1969)

<sup>11</sup> Guthrie, K.M., *Process Plant Estimating, Evaluation and Control*, Craftsman, Solano Beach, California (1974)

method f.o.b purchase cost of each piece of equipment is estimated as is the case with the Lang method. Instead of using an overall factor to account for installation of the equipment and other capital costs, individual factors for each type of equipment are used. This allows the materials of construction to be different for a reactor or separation unit and the platforms and ladders required to access it for example. To the summation of installed equipment costs, the components of total permanent investment including; contingency and contractor fee, site development costs, building costs and offsite facility costs, are added. The total permanent investment is added to the working capital to determine the total capital investment. The estimate uses the Chemical Engineering Cost Index to account for inflation by equipment type in this industry. The estimate is accurate to approximately  $\pm 20\%$ .

In all cases, the capital costs have been estimated by the Guthrie method where possible or the Lang method where not. The annualized cost of the capital investment are determined over the life of the plant giving an annualized capital expenses which is added to the annual operating costs for the plant to determine the annual cost for producing the annual production of the plant – 1.5 kg Pu-238. The annual cost divided by the annual production rate of the plant gives the supply cost for that year. Making assumptions about the sales price for the Pu-238 and its price sensitivity, the pre-tax profit from the production and upgrading operations developed will be determined as well as the depreciation, depletion and income taxes for these operations. Finally various rigorous profitability measures like annual cash flow, annual net present value and investor's rate of return will be determined for each scenario.

### **Capital Expenditures**

The Bare module cost of equipment for the Pu-238 chemical separation process is given in Table 3. Bare module cost consists of the fob purchase cost plus the cost of ancillary equipment required to completely install the equipment making it ready to use. Tank and mixer costs are determined from fob catalogue prices given at [www.indco.com](http://www.indco.com). Very small tanks ( 5 gal) are required for the ion exchange columns minimizing the radioactive materials inventory at any point in time. Pump costs were determined by the method of Lang. The cost of the RO unit was determined from a catalogue cost taken from <http://www.apswater.com/shopdisplayproducts.asp?id=749&cat=Commercial-Reverse-Osmosis-Systems>.

Table 3 Bare Module Cost of Equipment for Pu-238 Chemical Separation Process

Storage Tanks	Number	Size (gal)	Cp (each)	FBM	CPI Factor	C.BM
DI Water Storage Tank	1	1,000	\$ 4,991.00	4.16	1	\$ 20,762.56
Nitric Acid Storage Tank	1	500	\$ 5,806.75	4.16	1	\$ 24,156.08
Fe(II) sulfamate Storage Tank	1	100	\$ 3,070.50	4.16	1	\$ 12,773.28
Hydrazine Storage Tank	1	100	\$ 3,070.50	4.16	1	\$ 12,773.28
HCl Storage Tank	1	500	\$ 5,806.75	4.16	1	\$ 24,156.08
Mixer Motors	1	1 Hp	\$ 3,756.75	3.3	1	\$ 12,397.28
Feed Tank	1	100	\$ 3,070.50	4.16	1	\$ 12,773.28
Reactor Hold up	1	10	\$ 1,000.00	4.16	1	\$ 4,160.00
Pool Hold up	1	10	\$ 1,000.00	4.16	1	\$ 4,160.00
Resin Vessel-Step 1	5	5	\$ 377.00	4.16	1	\$ 7,841.60
Wash Tank-Step 2	1	10	\$ 495.00	4.16	1	\$ 2,059.20
Wash Tank-Step 3	1	10	\$ 495.00	4.16	1	\$ 2,059.20
Wash Tank-Step 4	1	10	\$ 495.00	4.16	1	\$ 2,059.20
Precondition Wash Tank-Step 5	1	20	\$ 589.00	4.16	1	\$ 2,450.24
RO Unit	2	1.4gpm	\$ 7,935.78	4.16	1	\$66,025.69
Sub Total						\$ 210,606.96
Pumps	18	10 gpm	\$ 3,352.75	3.3	1.1936	\$ 237,709.44
Capsule Filling Station						\$ 60,500.00
Capsule Opening Station						\$ 54,000.00
Nuclear Reactor						\$35,000,000.00
Security Fence Extension						\$ 200,000.00
Total CAPE X						\$ 35,762,816.40

In Table 3, we see that the vessels of various sizes, pumps and the capsule handling equipment account for a small fraction of the capital expenditures. The extension of a security fence on an existing Cat.-1 facility at INL and the nuclear reactor account for the lion's share of the capital expenses. Total Bare module costs with these expenses are \$35,762,816 and those without are \$562,816 - a substantial difference.

Using the Guthrie and Lang methods the bare module cost are used to determine other costs needed to calculate the total capital investment. These capital cost categories include the cost for utility plants including, electricity, water and steam plant for water distillation, site preparation costs, service facilities, contingencies and contractor fees, cost of land and plant startup. Working capital is estimated at 17.6% of total capital investment since the Np-237 and Pu-238 inventory must known to calculate this figure more accurately. As a result of this work the total capital investment is \$66 million. Given the cost estimation methods used this value is  $\pm 35\%$  assuming all major chemical separation units have been accounted for. The largest cost associated with the total capital investment is working capital accounting for more than 50% of the total capital investment. It should be noted that the site costs associated with an extension of an existing security fence is included in the site preparation costs associated with the total capital investment.

Table 4. Total Capital Investment

Allocated Cost for Utility		
Plants	\$	101,058
Site Preparation Costs	\$	1,614,422
Service Facilities	\$	2,152,563
Contingencies & Contractor		
fees	\$	1,937,307
Cost of Land	\$	331,363
Cost of Plant Startup	\$	1,658,117
Working Capital	\$	22,521,417
Total Bare Module Costs	\$	35,762,816
Total Capital Investment	\$	66,079,064

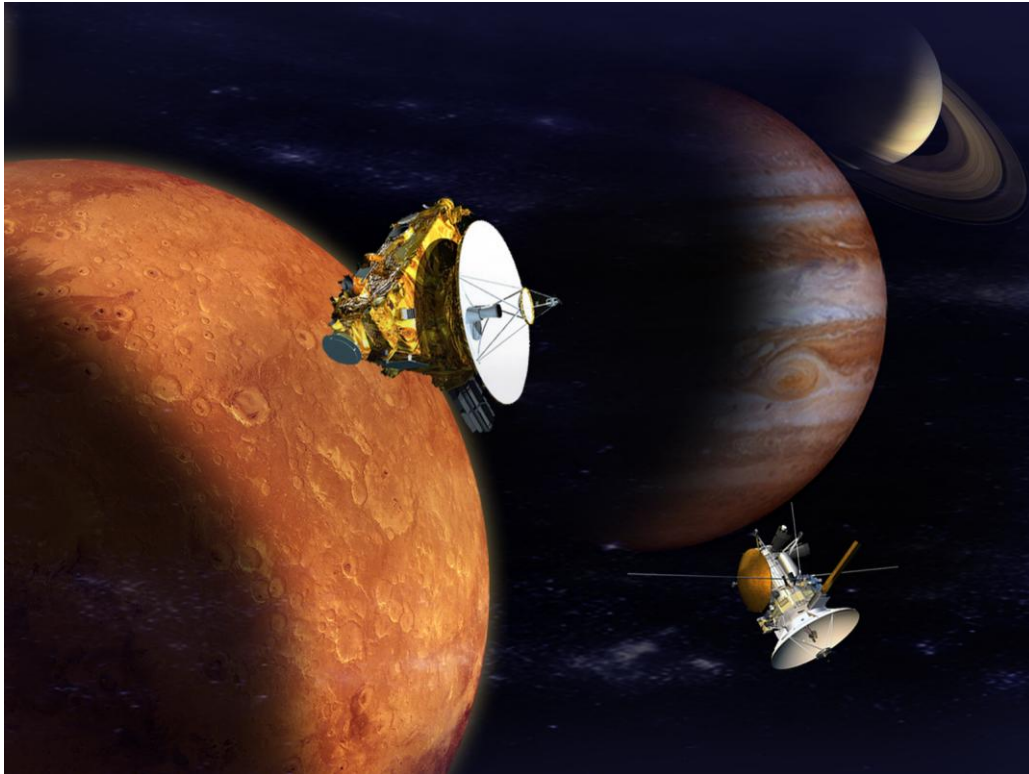
### Operating Expenses

From the prices of raw materials including \$200,000 per kg for Np-238, the cost of capital at 6.5% per year, cost of labor and waste disposal costs provide all of the annualized costs to produce Pu-238 on a per kg basis. As a result, the cost to separate Pu-238 in this process is \$3,245,104 per kg Pu-238. Labor costs in total account for \$1.26 million/yr half of which is required for security and half for nuclear/chemical operators. Thus, this process will be economically viable should the other expenses including the cost of operating the nuclear reactor and electroplating be less than \$2,000,000/yr as is likely the case.

Table 5 Operating Expenses

		Operational Expenses	
		\$ per kg Pu-238	
Cost of Capital @	6.50%	\$	1,780,093
Resin costs		\$	2,862
Np-237		\$	200,000
Steam		\$	0
Electricity		\$	3
Cooling Water		\$	91
Process Water		\$	16
Liquid Waste		\$	581
Labor Cost		\$	633,600
Security		\$	630,720
Process Costs	Total	\$	3,245,104 \$ per kg Pu-238
		+/- 50% estimate	
Sale Price		\$	6,000,000 \$ per kg Pu-238





The new method of production may enable a small, private reactor to produce sufficient quantities of Pu-238 to maintain the NASA outer planets exploration schedule.